Journal of Colloid and Interface Science **234,** 106–116 (2001)
doi:10.1006/jcis.2000.7301, available online at http://www.idealibrary.com on IDEAL®

Thermal Marangoni Convection of a Fluid Film Coating a Deformable Membrane

L. A. Dávalos-Orozco

Instituto de Investigaciones en Materiales, Universidad Nacional Autónoma de México, Ciudad Universitaria, Apartado Postal 70-360, Delegación Coyocán, 04510 México D.F., Mexico

Received April 12, 2000; accepted October 23, 2000

The thermal Marangoni instability of a fluid film coating a deformable membrane has been investigated by taking into account the deformation of the fluid free surface. Numerical calculations for different thermal boundary conditions are presented. The prestressed membrane is supposed to be very thin and therefore its behavior is similar to that of an isothermal fluid free surface with a surface tension but with a different mechanical boundary condition; that is, the fluid should stick on its surface and thus the fluid velocity is zero. An important assumption is that the membrane has no temperature dependence and therefore that only one Marangoni number exists for the free surface of the fluid. Numerical results are presented for stationary and oscillatory thermocapillary instability in both the sinuous and the varicose modes. It is shown that membrane deformation has important implications on the Marangoni instability of the fluid layer for positive and negative Marangoni numbers.

Key Words: **Marangoni convection; thermocapillary convection; membrane; surface deformation.**

I. INTRODUCTION

The problem of fluid motion due to temperature differences across a fluid film has important consequences in the finishing of coated surfaces. It is of interest to find out the temperature gradient necessary to destabilize the fluid layer in order to avoid any motion which may cause waves on the free surface. It has been found that these waves are related to the unevenness found after the coating film has solidified. Therefore, it is important to investigate the instability of the film under different thermal and mechanical boundary conditions. If the fluid layer is very thin buoyancy effects are not important and only thermocapillary or thermal Marangoni (that is, surface tension changes due to a temperature gradient) effects need be taken into account. The stationary case of this problem was first investigated by Pearson (1) for a lower flat wall and an upper free surface without deformation. A variety of mechanical and thermal boundary conditions were used. It was shown numerically by Vidal and Acrivos (2) that the marginal state for surface tension induced convection is stationary as assumed by Pearson (1).

Free surface deformation was taken into account for the first time by Scriven and Sternling (3) in the stationary case and for a lower flat solid wall. They also used different mechanical and thermal boundary conditions. As shown for the flat free surface by Vidal and Acrivos (2), it was shown numerically by Castillo and Velarde (4) that the Marangoni number of the marginal state of stationary convection is smaller than the corresponding one for oscillatory convection in the presence of surface deformation. That is, stationary convection appears first.

Funada (5) investigated the thermocapillary instability of a static liquid sheet, that is, a thin liquid layer which has two free deformable surfaces that are both susceptible to temperature gradients. In this case, two Marangoni numbers are necessary to describe the instability of the sheet. Funada investigated two cases typical in the theory of liquid sheets, that is, the sinuous and the varicose deformation modes. In the first one the waves on the free surfaces are in phase and in the second one they are out of phase by 180°.

Dávalos-Orozco (6) investigated the thermocapillary instability of a liquid sheet in motion from the point of view of the most unstable mode. There, it was shown how the sinuous deformation mode may be the most unstable in the presence of a large enough temperature gradient. Note that the sinuous and varicose modes of instability also may appear before the rupture of isothermal thin liquid films (7).

In this paper, the thermal Marangoni instability of a thin fluid layer coating a deformable prestressed membrane is investigated. The idea is to investigate, in a simple way, the effect a deformable wall may have on the thermocapillary instability in comparison to classical results found in earlier papers (1–4). The membrane is supposed to be very thin but with a prescribed tension. It is assumed that this tension does not change due to temperature gradients and therefore behaves as an isothermal free liquid surface. However, the important difference it has with respect to a free surface is that the fluid coating the membrane sticks to it due to viscous effects and thus the nonslip boundary condition is applied to make the velocity zero on it. This assumption is what makes the difference between the results of the paper by Funada (5) and those of the present one. Here, only one Marangoni number, that of the free surface, describes the thermocapillary instability but again two crispation numbers are

0021-9797/01 $35.00

available to account for the relative inverse tensions of the free surface and the membrane. These crispation numbers also represent the ability of the free surface and the membrane to deform.

Note that the membrane is the simplest model an elastic wall may have as boundary condition for a fluid system. The results given herein will show that deformable boundaries have in fact important consequences on the instability of coating fluid layers and in particular on their thermocapillary convection instability.

The structure of the paper is as follows. In Section II the equations of motion and boundary conditions are presented. In Section III the stationary and oscillatory instabilities are investigated. Section IV is the discussion and conclusions.

II. EQUATIONS OF MOTION

We investigate the Marangoni instability of a thin liquid layer coating a deformable prestressed membrane. The physical system is sketched in Fig. 1. In the figure it is seen that the membrane is located at $z = 0$ and the fluid free surface is located at $z = d$. The membrane is presented as being hotter than the free surface but, as will be shown presently, instability may also occur when the situation is reversed, that is, when the layer is heated from above.

Two kinds of instability are investigated as shown in Fig. 2. They are the sinuous and varicose deformation modes. In the sinuous mode both the membrane and the free surface are deformed in phase. In the varicose mode the membrane and free surface are out of phase by 180°. The stabilities of these two modes were also investigated by Funada (5) and Dávalos-Orozco (6) under different conditions.

The equations governing the flow of the system are the following. The balance of inertial forces with the pressure and viscous forces is expressed by the Navier–Stokes equation,

$$\rho\left[\frac{\partial \vec{u}}{\partial t} + \vec{u}\cdot\nabla\vec{u}\right] = -\nabla p + \mu\nabla^2\vec{u}. \qquad [1]$$

The continuity of the velocity is expressed by the incompressibility condition,

$$\nabla\cdot\vec{u} = 0. \qquad [2]$$

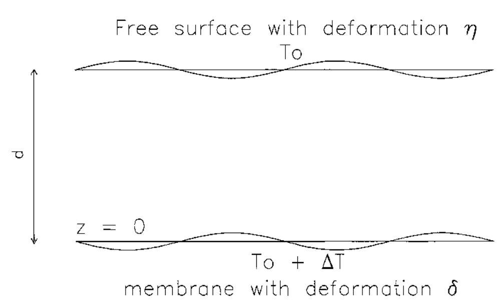

FIG. 1. Sketch of the system under research. It is composed of a free deformable surface and a deformable membrane. The fluid layer has an imposed temperature gradient.

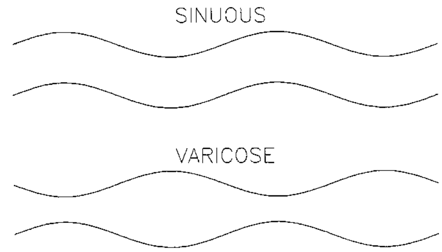

FIG. 2. Two modes of deformation investigated: The sinuous and the varicose modes.

The heat transfer is described by the diffusion equation for the temperature,

$$\frac{\partial T}{\partial t} + \vec{u}\cdot\nabla T = \kappa\nabla^2 T, \qquad [3]$$

where $\vec{u}$ is the velocity vector which in two dimensions has components (u, w), p is the pressure, ρ is the density, μ is the viscosity, T is the temperature of the fluid layer, and κ is the heat diffusivity.

The boundary conditions of the problem are as follows. At the membrane, located at $z = \delta$, where δ is the membrane deformation displacement from its equilibrium position at $z = 0$, the conditions are that the fluid should stick to the surface due to friction, that is,

$$u = 0. \qquad [4]$$

The membrane follows the fluid motion and therefore satisfies the kinematic boundary condition,

$$w = \frac{\partial \delta}{\partial t}, \qquad [5]$$

where use has been made of the condition Eq. [4] and consequently the missing term $u\partial\delta/\partial x$ is zero. The heat transfer through the thin membrane from the fluid to the atmosphere is given by

$$k\nabla T\cdot\vec{n}_\delta = -q_\delta(T - T_0 - \Delta T). \qquad [6]$$

The stress jumps at the membrane are related to its curvature and to its tension σ_δ by

$$\left(S^f_{ij} - S^a_{ij}\right)\cdot n_{\delta i} = n_{\delta j}K_\delta\sigma_\delta \qquad [7]$$

$$\vec{n}_\delta = (\delta_x, -1)/N_\delta. \qquad [8]$$

At the free surface, located at $z = 1 + \eta$, where η is the free surface deformation from the equilibrium position at $z = 1$, the kinematic boundary condition is

$$w = \frac{\partial \eta}{\partial t} + u\frac{\partial \eta}{\partial x}. \qquad [9]$$

The heat transfer through the free surface is expressed by

$$k\nabla T \cdot \vec{n}_\eta = -q_\eta(T - T_0). \quad [10]$$

The stress jumps due to free surface curvature, surface tension σ, and thermocapillary effects are given by

$$\left(S_{ij}^f - S_{ij}^a\right) \cdot n_{\eta i} = n_{\eta j} K_\eta \sigma - \left(-\frac{d\sigma}{dT}\right)(\nabla T \cdot \vec{t}_\eta) t_{\eta j} \quad [11]$$

$$\vec{n}_\eta = (-\eta_x, 1)/N_\eta, \quad \vec{t}_\eta = (1, \eta_x)/N_\eta, \quad [12]$$

where the condition for the normal stresses is found by multiplying by the vector normal to the free surface $\vec{n}_\eta$ and the condition for the tangential stresses is obtained by multiplying by the tangential vector $\vec{t}_\eta$. The subindexes of δ and η indicate the partial derivative in x.

Here, k is the thermal conductivity of the fluid and q_δ and q_η are the heat transfer coefficients of the membrane and the free surface, respectively. $K_\delta = \delta_{xx}/N_\delta^3$ and $K_\eta = \eta_{xx}/N_\eta^3$ are the curvatures of the membrane and the free surface, respectively, and $N_\delta = (1 + \delta_x^2)^{1/2}$, $N_\eta = (1 + \eta_x^2)^{1/2}$. The stresses of the fluid and the air are $S_{ij}^f = -p\delta_{ij} + \tau_{ij}^f$ and $S_{ij}^a = -p\delta_{ij}$, respectively where δ_{ij} is the Kronecker delta. The term τ_{ij}^f is the viscous stress tensor of a Newtonian fluid. The viscous stresses of air are neglected and only its pressure is taken into account. In the case of the membrane only the normal stresses are needed because use has been made of the stick boundary condition Eq. [4]. Note that $-d\sigma/dT$ is positive for almost all fluids.

The above equations of motion are linearized after giving a perturbation to the variables. The linear equations of motion of the perturbation are those for a fluid layer heated from below in the absence of gravity and are the same as those used by Pearson (1) and Scriven and Sternling (3). The differences with our problem appear at the boundary conditions. The boundary conditions must be expanded in Taylor series of δ and η around $z = 0$ for the membrane and $z = 1$ for the free surface, respectively.

The variables are made nondimensional by means of the thickness of the layer d for length, d^2/κ for time, κ/d for velocity, $\rho_o \nu\kappa/d^2$ for pressure and stresses, and ΔT for temperature. Here, ρ_o is a reference density, ν is the kinematic viscosity, and ΔT is the temperature difference between the membrane and the free surface, which is at a reference temperature T_o. The time and velocity have been made nondimensional by using κ because the thermal diffusivity determines the magnitude of time and speed of heat conduction from the lower boundary to the upper one which deforms with a change in temperature affecting the strength of the resulting motion after the perturbation. In this way, after nondimensionalization, the thermal diffusivity appears in the term of the Navier–Stokes equation containing the Prandtl number which measures the relative importance of viscosity and thermal diffusivity. Note that under hydrostatic conditions the nondimensional temperature profile of the basic state is $\overline{T} = -z + 1 + T_0/\Delta T$.

The linear equations are combined, eliminating the pressure, to obtain two equations for the vertical component of the velocity perturbation w and for the temperature perturbation θ. The solutions of those equations are obtained by separating variables by means of normal modes of the form $(w, \theta, \delta, \eta) = (W, \Theta, \Delta, H) \exp i(ax - \omega t)$ where $i = \sqrt{-1}$, a is the wavenumber, and ω is the frequency. Upon substitution, the equations of motion and heat transfer are

$$(D^2 - a^2)\left(D^2 - a^2 + i\frac{\omega}{Pr}\right)W = 0 \quad [13]$$

$$(D^2 - a^2 + i\omega)\Theta = -W, \quad [14]$$

where W and Θ are the amplitudes of the z-component of the velocity and the temperature, respectively, and $D = d/dz$. Δ and H are the amplitudes of the membrane and surface deformations, respectively.

The boundary conditions in normal modes and in nondimensional form are
At the deformable membrane, at $z = 0$:

$$W = -i\omega\Delta \quad [15]$$

$$DW = 0 \quad [16]$$

$$D\Theta = B_\delta(\Theta - \Delta) \quad [17]$$

$$\left(\frac{i\omega}{Pr} + D^2 - 3a^2\right)DW = \frac{a^4}{C_\delta}\Delta. \quad [18]$$

At the free surface, at $z = 1$:

$$W = -i\omega H \quad [19]$$

$$D\Theta = -B_\eta(\Theta - H) \quad [20]$$

$$\left(\frac{i\omega}{Pr} + D^2 - 3a^2\right)DW = \frac{a^4}{C_\eta}H \quad [21]$$

$$(D^2 + a^2)W = -Ma^2(\Theta - H). \quad [22]$$

Note that the amplitudes Δ and H in Eqs. [17], [20], and [22] appear due to linearization after taking a Taylor expansion of temperature around $z = 0$ and 1, respectively.

The nondimensional parameters which appear in the equations and boundary conditions are defined as follows. $Pr = \nu/\kappa$ is the Prandtl number. $B_\delta = q_\delta d/k$ and $B_\eta = q_\eta d/k$ are the Biot numbers of the membrane and free surface, respectively. $C_\delta = \rho_o \nu\kappa/\sigma_\delta d$ and $C_\eta = \rho_o \nu\kappa/\sigma d$ are the crispation numbers of the membrane and free surface, respectively. The Marangoni number at the free surface is $M = \gamma \Delta T d/\rho_o \nu\kappa$, where $\gamma = -d\sigma/dT$ is positive.

In the next section the results of the numerical calculations are presented. The stationary convection is considered before oscillatory convection.

III. STATIONARY AND OSCILLATORY SOLUTIONS

The boundary value problem represented by the system of equations given above can be solved analytically as in the cases of Refs. (1, 3, 5, 6). After obtaining a large formula for M it is necessary to calculate its value numerically because of the difficulty in obtaining some physical insight into the terms. However, simple analytical formulas will be given when an adequate limit process is possible.

A. Stationary Marangoni Convection

The formula obtained for M in the case of stationary Marangoni convection includes all the parameters involved in the problem. Thus, M is

$$M = \frac{8a(a - \sinh a \cosh a)[B_\delta(a + B_\eta \tanh a) + a(B_\eta + a \tanh a)]}{B_\delta M_1 + a M_2}. \quad [23]$$

Here, M_1 and M_2 are

$$M_1 = a^3 - \tanh a \sinh^2 a + 8C_\delta a^2 \tanh a - 8C_\eta a^3 \quad [24]$$

$$M_2 = a^3 \tanh a - a^2 + 2a \tanh a - \sinh^2 a - 8C_\eta a^3 \tanh a. \quad [25]$$

This equation is simpler than that for the oscillatory case. Note that a change of sign in front of C_δ corresponds to the varicose mode.

Some particular cases are found in which M reduces to the previous results:

(1) When $C_\delta = C_\eta = 0$ the problem reduces to that of Pearson (1) for a solid wall and a free nondeformable surface.

(2) When $C_\delta = 0$ the problem reduces to that of Scriven and Sternling (3) for a solid wall and a free deformable surface.

(3) When $B_\delta = 0$ the problem reduces to that of Scriven and Sternling (3) for the same Biot number. This means that the wall or membrane deformation has no influence on the stationary instability when the heat flux passing through it is fixed.

There are also some analytical results obtained when the limit $a \to 0$ is taken in Eq. [23]. The general formula in this limit is

$$M = \frac{2}{3}\frac{B_\delta(1 + B_\eta) + B_\eta}{B_\delta(C_\eta \pm C_\delta)}a^2. \quad [26]$$

Note that the plus sign corresponds to the varicose mode and the minus sign to the sinuous mode. This Marangoni number of the sinuous mode is positive when $C_\delta \le C_\eta$, is negative when the inequality is reversed, and tends to zero when $a \to 0$.

Another interesting limit of Eq. [23] when $a \to 0$ is obtained only for the sinuous mode when the two crispation numbers are equal, that is, $C_\eta = C_\delta$. Under these conditions the Marangoni number for the sinuous mode,

$$M = 2\frac{B_\delta(1 + B_\eta) + B_\eta}{C_\eta(3 + B_\delta)}, \quad [27]$$

has a finite value. This Marangoni number of the sinuous mode has no counterpart in previous papers because both the membrane and the free surface have deformations. When the membrane is a very good conductor, $B_\delta \to \infty$, the Marangoni number in Eq. [27] tends to $M = 2(1 + B_\eta)/C_\eta$.

In addition, when $C_\eta = C_\delta$ the result for the varicose mode may be obtained directly from Eq. [26] using the plus sign. Therefore, M for the varicose mode also tends to zero as $a \to 0$.

As explained above, the membrane deformation has no influence on the stability when $B_\delta = 0$. In this case, the limit $a \to 0$ gives

$$M = \frac{2}{3}\frac{B_\eta}{C_\eta}, \quad [28]$$

a finite value for M, and when $B_\delta = B_\eta = 0$ the limit is

$$M = \frac{2}{3}\frac{1}{C_\eta}a^2, \quad [29]$$

where M tends to zero. Here, both Eqs. [28] and [29] are the same as those obtained by Scriven and Sternling (3) under the same conditions.

The numerical results for the marginal curves of the Marangoni number against the wavenumber will be presented in what follows. First, results for the varicose mode and then those of the sinuous mode will be given. Two values, $B_\delta = 1$ and ∞, were investigated for each mode with variation of the magnitudes of the other parameters.

1. Varicose mode. The varicose mode has two interesting properties. One is that the calculated marginal curves of the Marangoni number against the wavenumber are always smaller (more instability) than those corresponding to the flat wall. The marginal Marangoni numbers of the flat wall were calculated again to be able to compare them numerically with our new results. The second property is that the varicose mode also has smaller values of the marginal Marangoni number compared to those of the sinuous mode. The results of the sinuous mode are presented for reasons which will be explained presently.

In general, the behavior of the marginal curves is similar to that of the flat case (3) as seen in Fig. 3 for $B_\delta = 1$. Figure 3a for a strong membrane tension with $C_\delta = 0.0001$ shows how all the curves of M tend to zero as a tends to zero. However, for C_η smaller than 0.01 local minima appear for all the values of B_η calculated. A comparison of our numerical results with those of the flat wall shows that for small wavenumbers M may have half of the value of the flat case. When the wavenumber increases both values approach each other but with the M of the varicose mode always smaller.

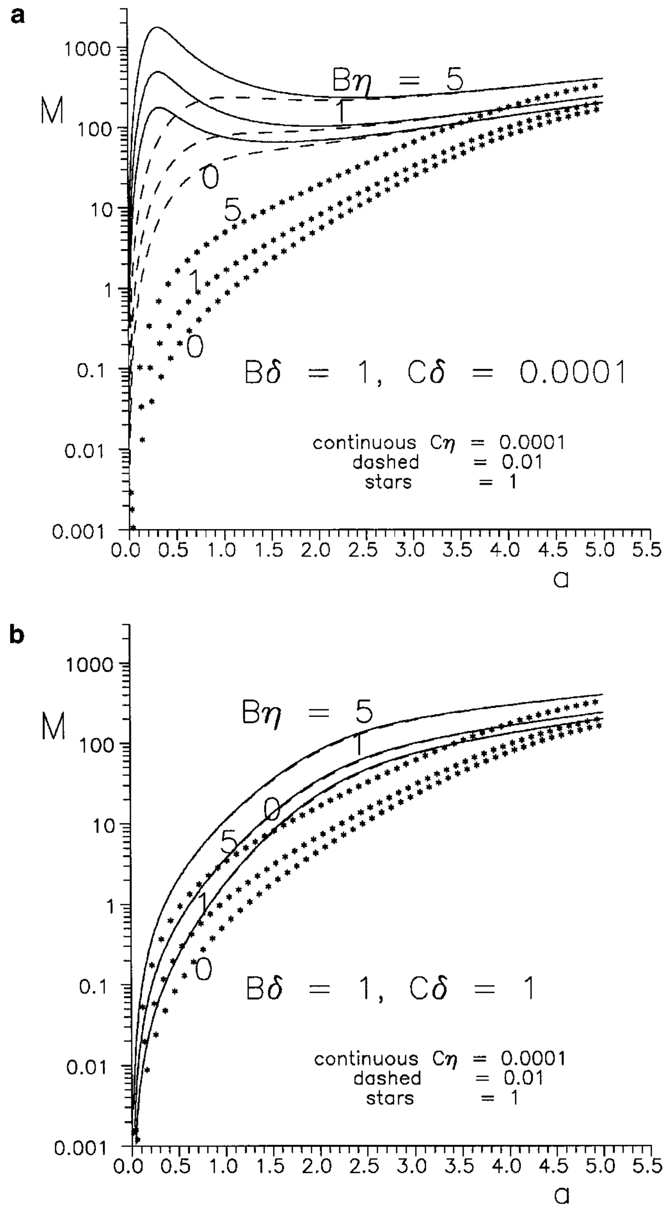

FIG. 3. Stationary convection, varicose mode. Marginal curves of the Marangoni number against the wavenumber for $B_\delta = 1$, three different values of C_η, and two different values of C_δ: (a) $C_\delta = 0.0001$, (b) $C_\delta = 1$.

Calculations were made also for $C_\delta = 0.01$. The behavior of the curves is similar but it seems that the local minima begin to be undetectable with increasing C_δ. The minima already disappear in Fig. 3b for $C_\delta = 1$.

Note that the starred curves ($C_\eta = 1$) are almost the same for the values of C_δ in Figs. 3a and 3b. This is the origin of the interesting result that, when $C_\delta = 1$ (small membrane tension) in Fig. 3b and the wavenumbers are smaller than a certain value, the fluid layer with $C_\eta = 1$ (small surface tension) is more stable than other fluids with stronger surface tension.

The behaviors of the curves of Fig. 4 seem to be similar to those of Fig. 3. However, a detailed comparison of them leads to interesting conclusions. For wavenumbers lower than a certain value and depending on the magnitude of C_η the curves for $B_\delta = \infty$ are lower (more instability) than those of $B_\delta = 1$. For example, in Fig. 4a the curves corresponding to $C_\eta = 0.01$ and $B_\eta = 0$ are smaller than those of Fig. 3a below a wavenumber $a \leq 0.09$ where they intersect, and for $B_\eta = 1$ below an intersection $a \leq 0.83$. Here, the value of a at which the curves cross each other will be called the intersection wavenumber. Above that point the curves behave as expected; that is, the system is more unstable when the wall Biot number B_δ is smaller because it is more difficult for the perturbation to leave the fluid layer through the membrane. However, it seems that the membrane

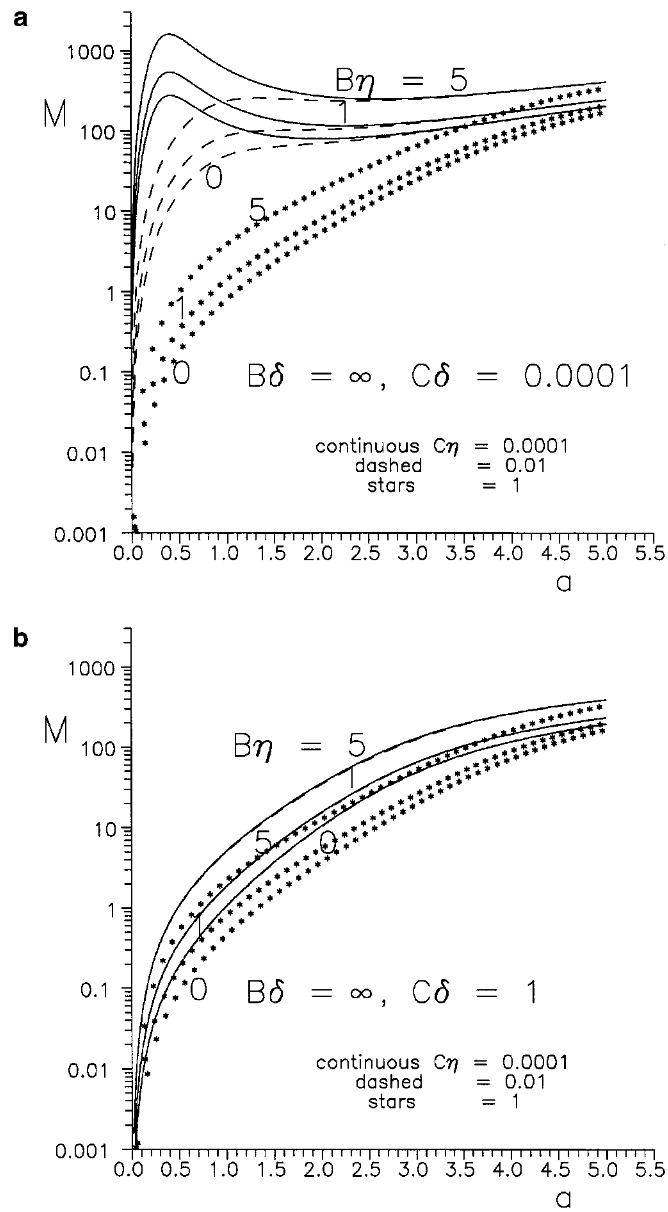

FIG. 4. Stationary convection, varicose mode. Marginal curves of the Marangoni number against the wavenumber for $B_\delta = \infty$, three different values of C_η, and two different values of C_δ: (a) $C_\delta = 0.0001$, (b) $C_\delta = 1$.

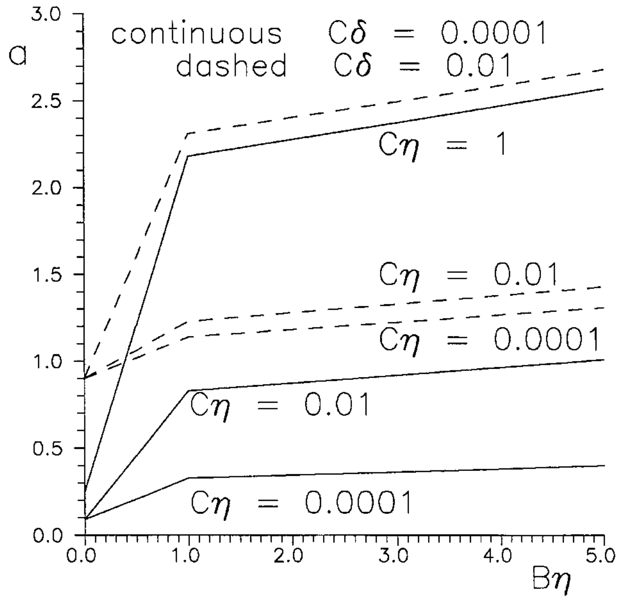

FIG. 5. Stationary convection, varicose mode. Curves of the intersection wavenumber a against B_η, the free surface Biot number. They show the value of a below which the marginal curves corresponding to $B_\delta = \infty$ are lower (more instability) than those of $B_\delta = 1$.

deformation contributes to the instability of the fluid layer and makes it more unstable for very large Biot number and small enough wavenumbers. See Fig. 5 for plots of the intersection wavenumber against B_η, corresponding to the results shown in Figs. 3 and 4 and for $C_\delta = 0.01$.

In Fig. 5 the results for $C_\delta = 1$ are not presented. Note that the curves in Fig. 4b show that the fluid layer is more unstable for $B_\delta = \infty$ than for $B_\delta = 1$ in the entire range of wavenumbers presented (but checked until $a = 15$). In fact, if all the numerical tables of the curves of Figs. 3b and 4b are compared it may be seen that no crossing occurs (in the range $0.01 \leq a \leq 15$) but it is found that the corresponding curves in both figures tend to the same magnitude (within four decimals) for a very large a. Figure 5 shows that the intersection wavenumber increases with B_η and C_δ. As explained above, a tends to very large values (no intersection) when $C_\delta = 1$.

Note that the intersection of the curves occurs also for other values of B_δ. However, the reason this intersection exists has not been discussed. As explained above, when $B_\delta = 0$ the stationary problem reduces to that of the flat wall which is more stable than the results given here indicate. Therefore, when the membrane Biot number is greater than zero the marginal curves are lower than those of the flat wall due to the new degrees of freedom given by the membrane to the fluid layer. This effect is stronger when B_δ is very large and the marginal Marangoni numbers can be smaller but, as can be seen by comparison of Figs. 3 and 4, this result is only allowed below certain intersection wavenumbers, as shown in Fig. 5.

As discussed above, Eq. [26] was obtained by an expansion of M in terms of very small wavenumbers. By means of this equation it may be observed that in the limit of very small a the Marangoni number for $B_\delta = 1$ is again larger than that for $B_\delta = \infty$. This could not be detected numerically in this limit. Consequently, this equation confirms our results. The finite limit found in Eq. [28] supports the numerical result which shows that for $B_\delta > 0$ the Marangoni number in Eq. [26] is smaller (tends to zero) than that when B_δ is zero.

2. Sinuous mode. It is found that the sinuous mode has larger values of the marginal Marangoni numbers (more stable) than the varicose mode. However, this only occurs when M is positive. It will be shown presently that the sinuous mode is also unstable to negative Marangoni numbers. In the varicose mode this property is absent and therefore only the sinuous mode will appear when M is negative and large enough for a given wavenumber.

The negative Marangoni number appears here as a result of the deformation of the membrane which gives an extra degree of freedom to the motion of the fluid layer. If a perturbation is given to the free surface when it is heated from above, the free surface moves and the perturbation is transmitted to the membrane which only opposes the motion by friction and by the degree to which its tension increases. However, depending on the magnitude of the wavenumber this opposition may or may not be important. For relatively small wavenumbers not only viscous but also surface tension and tension effects are not very important and the membrane can follow the free surface perturbation stimulating its growth. Therefore, the perturbation is not dissipated like in a flat wall. From the results given below, it seems that for negative Marangoni numbers and relatively small wavenumbers the easiest way for the free surface and the membrane to move along is by means of the sinuous mode.

The case for $B_\delta = 1$ of the stationary sinuous mode is presented in Fig. 6. Figure 6a shows the marginal curves when $C_\delta = 0.0001$ and for three different values of C_η. The continuous curves corresponding to $C_\eta = 0.0001$ have a very large finite limit value when a is small as expected from Eq. [27]. In the curves for other B_η the Marangoni number tends to zero as $a \to 0$. Notice that the curves for $C_\eta = 0.0001$ and $B_\eta = 1$ to 5 have a local minimum at a finite value of a, which is still present when $C_\eta = 0.01$ and $B_\eta = 5$.

The results corresponding to the value of $C_\delta = 1$ are plotted in Fig. 6b. In this figure it is shown that a singular value of a appears where the denominator of M given in Eq. [23] becomes zero and changes sign. For $C_\eta = 0.0001$ and 0.01 this particular value is around $a = 2.3$. At this point the value of the Marangoni number changes from negative to positive when a is increased from zero. The negative Marangoni numbers have very large values when $a < 2.3$ and tends to zero from below as $a \to 0$. When the wavenumber is larger than the singular one the marginal M drops from a very large positive value to a local minimum for a finite value of the wavenumber and then increases again smoothly. The starred curves of $C_\eta = 1$ have a finite limit as $a \to 0$ and also have a minimum for a value of the wavenumber which is zero in this particular case.

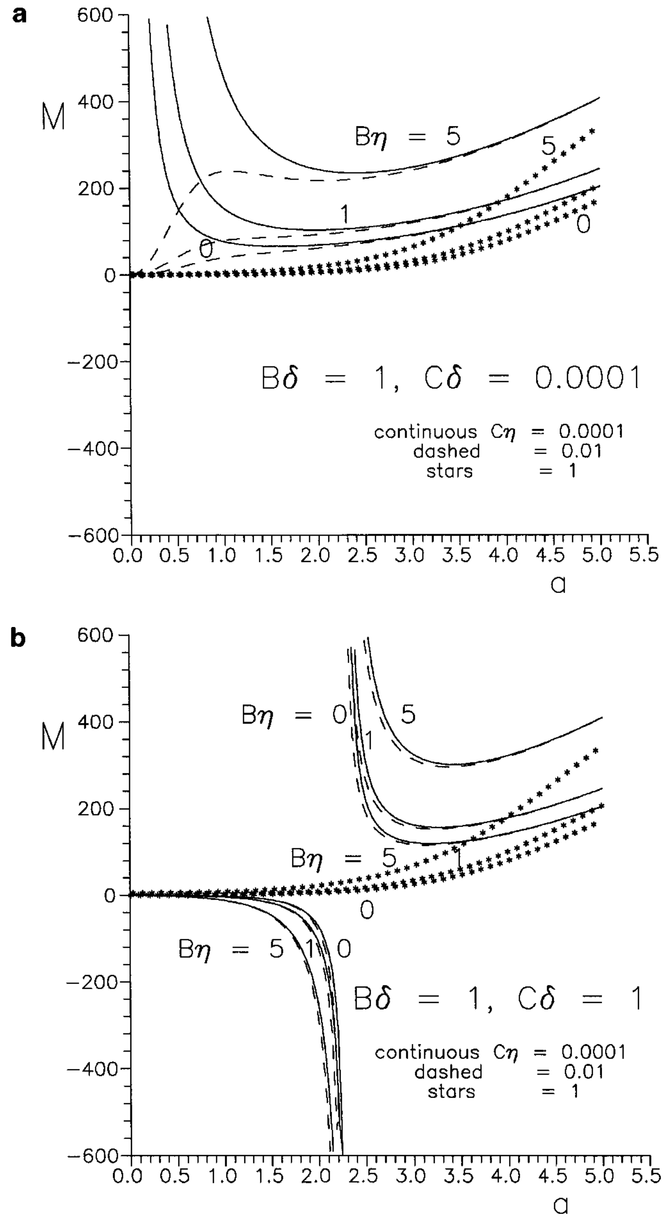

FIG. 6. Stationary convection, sinuous mode. Marginal curves of the Marangoni number against the wavenumber for $B_\delta = 1$, three different values of C_η, and two different values of C_δ: (a) $C_\delta = 0.0001$, (b) $C_\delta = 1$.

Calculations were made also for of $C_\delta = 0.01$. It has been shown that only the curves of $C_\eta = 0.0001$ have a singularity and therefore have negative values of the Marangoni numbers for wavenumbers smaller than a singular one around $a = 0.8$. To the right they also show a minimum for finite values of the wavenumber. Only the curves for $C_\eta = 0.01$ have limits of M different from zero as $a \to 0$ and their minima are at a finite value of a.

In Fig. 7 the value of the Biot number of the membrane is changed into that of a very good conductor, $B_\delta = \infty$. The behavior of the curves in Fig. 7 is similar to that found in Fig. 6. However, it is interesting to see how the singularity values of the wavenumber have been displaced to the right with respect to the previous figure.

In Fig. 7a it is found again that the curves are smaller than those corresponding in Fig. 6a for wavenumbers smaller than a certain magnitude, as discussed in Figs. 3 and 4. Exception is made for the continuous curves of $C_\eta = 0.0001$. For the larger magnitude of C_δ shown in Fig. 7b the behavior of the curves above the a of singularity is as expected. However, below the singular a it is found that the negative curves are smaller in magnitude for $B_\delta = \infty$ than for $B_\delta = 1$. This means that the marginal curves of the negative Marangoni number are more unstable when the

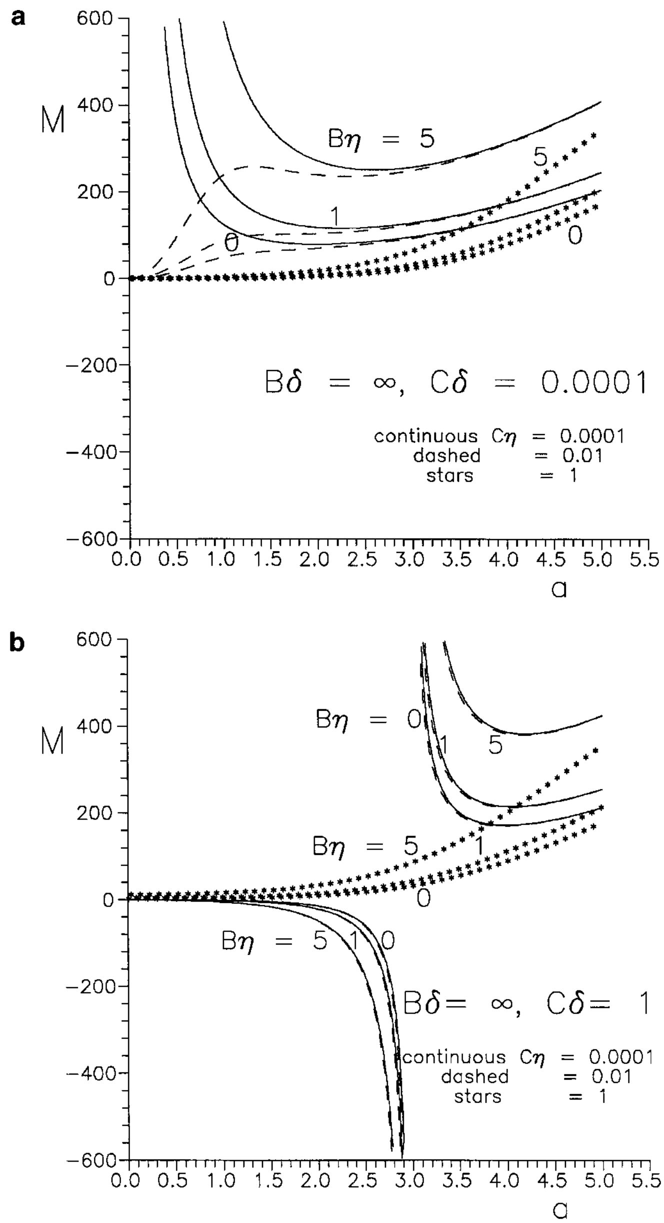

FIG. 7. Stationary convection, sinuous mode. Marginal curves of the Marangoni number against the wavenumber for $B_\delta = \infty$, three different values of C_η, and two different values of C_δ: (a) $C_\delta = 0.0001$, (b) $C_\delta = 1$.

TABLE 1

Comparison of the Marginal Marangoni Numbers against Wavenumber *a* (First Column) for the Stationary Case of a Flat Wall (Scriven and Sternling, 1964, Second and Ninth Columns, S&S), Oscillatory Case for a Flat Wall (Castillo and Velarde, 1982, Third and Tenth Columns), Oscillatory Sinuous Case of a Deformable Membrane (Fourth and Fifth Columns), and Oscillatory Varicose (Var) Case of a Deformable Membrane (Sixth Column)

	$Pr = 7$, $B_\delta = B_\eta = 0$															
	$C_\delta = 0$, $C_\eta = 0.0001$		$C_\delta = 1$, $C_\eta = 0.0001$			$C_\delta = 0.001$, $C_\eta = 0.0001$			$C_\delta = 0$, $C_\eta = 0.01$		$C_\delta = 1$, $C_\eta = 0.01$			$C_\delta = 0.001$, $C_\eta = 0.01$		
			Sinuous			Sinuous					Sinuous			Sinuous		
	Stat	Flat			Var			Var	Stat	Flat			Var			Var
	S&S	Osc.	Osc.	Osc.	Osc.	Osc.	Osc.	Osc.	S&S	Osc.	Osc.	Osc.	Osc.	Osc.	Osc.	Osc.
a	M	M	M1	M2	M	M1	M2	M	M	M	M1	M2	M	M1	M2	M
0.01	0.584	—	0.375	−1.94	0.029	0.385	−2.98	0.048	0.007	—	0.003	−1.87	0.028	0.014	−233	0.048
0.02	2.730	—	1.502	−2.38	0.097	1.481	−11.1	0.194	0.027	—	0.018	−2.37	0.096	0.056	−232	0.192
0.05	12.47	14.32	9.862	−3.06	0.341	10.08	−20.8	1.234	0.166	—	0.118	−3.25	0.115	0.368	−277	1.230
0.065																2.143
0.1	27.97	51.60	38.84	−4.31	0.772	39.31	−40.0	21.91	0.658	—	0.469	−4.33	0.470	1.570	−218	—
0.2	40.82	201.2	143.3	−6.35	1.454	142.7	−123	57.21	2.545	—	1.879	−6.06	1.876	6.797	−207	—
0.3	44.75	397.9	290.0	−8.27	1.988	286.1	−926	132.1	5.422	6.36	4.196	−7.66	1.995	16.96	−209	—
0.4	46.47	562.1	461.8	−10.4	2.394	456.0	−7515	237.3	8.975	10.25	7.406	−9.42	2.402	33.88	−218	—
0.5	47.49	763.0	650.2	−13.4	2.672	648.8	−6897	303.3	12.89	15.41	11.49	−11.6	2.686	59.57	−231	—
0.6	48.25	1010	850.6	−17.5	2.800	864.3	−6377	292.4	16.92	21.94	16.45	−14.5	2.826	96.25	−250	—
0.7	48.95	1308	1060	−24.3	2.749	1103	−6214	279.6	20.87	29.92	22.27	−18.9	2.783	146.9	−273	—
0.8	49.64	1656	1277	−37.2	2.459	1366	−6396	407.7	24.65	39.40	28.97	−26.3	2.500	215.5	−301	—
0.9	50.35	2047	1500	−70.7	1.857	1654	−6897	654.1	28.20	50.26	36.54	−40.9	1.897	307.8	−335	—
1.0	51.13	2462	1728	−287	0.835	1967	−7765	1158	31.52	62.25	45.00	−81.4	0.873	432.1	−377	—
1.1	51.97	2861	1959	233.9	−0.59	2308	−9137	2534	34.61	75.17	54.38	−382	−0.51	601.3	−428	—
1.3									40.27	104.3	75.89	116.3	−3.68	1174	−567	
1.4	55.04	3597	2672	97.34	−2.23	3505	−21177	−2918								—
1.5	56.27	3745	2915	106.8	−6.20	3970	−34818	−2214	45.44	139.1	101.1	114.1	−7.46	2563	−781	—

Note: The values of the crispation numbers C_δ and C_η are as shown above the columns. To $C_\delta = 0$ corresponds two sets of two columns each. The other columns for $C_\delta > 0$ (from the 7th to 9th and from the 12th to 17th columns) show results for the values of the crispation numbers shown above them. The sinuous mode always shows two marginal curves. Blanks mean that no calculations were made. Dashes mean that no value was found.

membrane is a very good conductor than when it is a poor one. This is confirmed by Eq. [26].

B. *Oscillatory Marangoni Convection*

The oscillatory thermocapillary convection is investigated here. It is supposed that the Biot numbers of both the membrane and the free surface are zero. This means that the heat flux is fixed in both boundaries. Note again that when $B_\delta = 0$ the membrane deformation has no importance in the stationary case. Therefore, the marginal Marangoni numbers of oscillatory thermocapillary convection calculated here will be compared with the results of the stationary problem investigated by Scriven and Sternling (1) for a lower rigid flat wall and with the results of Castillo and Velarde (4) for oscillatory convection with a lower flat wall. No numerical data are available from these papers because they only show graphs of M against a. Consequently, all data were calculated numerically again for the sake of comparison.

The equation for the explicit analytical expression of the complex Marangoni number is far larger than that for the stationary case. Therefore, the result will not be given here. The numerical method to calculate the marginal Marangoni number against the wavenumber involves looking for the frequency which makes the imaginary part of M equal to zero for a fixed a. When two values are obtained for different frequencies where both M's are positive, that which corresponds to a smaller M is selected. If the two M's have different sign both are selected as physically possible.

The numerical results are presented in Table 1 along with a discusion of the consequences the change of parameters have on them. A table is an easy way to show the difference among very similar quantities. That difference is important to determine, from the stability point of view, if the stationary or the oscillatory convection will be the first unstable mode when the Marangoni number is increased for a fixed wavenumber. In addition, it will clearly show if the wall or membrane deformation contributes to destabilize even more the liquid layer in comparison with the well-known results published in the literature for a nondeformable flat wall.

Table 1 shows results for a variety of situations when $Pr = 7$. It is divided in two parts. In the first one results are shown for $C_\eta = 0.0001$ and in the second one for $C_\eta = 0.01$. In the first part calculations were made for C_δ less than or equal to C_η. In the second one, calculations were made for C_δ less than, equal to, and greater than C_η. The first column corresponds to the wavenumber a, the second and tenth columns to the marginal Marangoni number of the stationary case with free surface deformation and flat wall (3), and the third and eleventh columns to M

of the oscillatory case with surface deformation and flat wall (4), respectively. The following columns are separated in sets of three columns each. The first set of the first part of Table 1 corresponds to $C_\delta = 1$ and the second to $C_\delta = 0.001$, respectively, but results were obtained also for $C_\delta = 0.01$ and $C_\delta = 0.0001$. The same explanation corresponds to the second part of Table 1, starting from column 12. The first two columns of each set correspond to the sinuous mode and the third one to the varicose mode, respectively. In Table 1 the blanks mean that the calculations were not made and the dashes mean that no marginal Marangoni number was found; that is, the imaginary part of M never changed sign over a very wide range of frequencies investigated.

From the second, third, tenth, and eleventh columns of Table 1 it is easy to see that the results of Castillo and Velarde (4) are correct; that is, for a flat wall the stationary convection occurs first. Thus, under these conditions it is of interest to know if the membrane deformation may contribute to destabilize the liquid layer more than in the stationary case of the flat wall (3), or in other words, if the Marangoni number might be smaller. Note that in the third column, for wavenumbers below $a = 0.05$, and in the eleventh column, for wavenumbers $a \leq 0.2$, it was not possible to find a change of sign of the imaginary part of the Marangoni number. Therefore, no oscillatory convection is possible from $a \to 0$ up to a wavenumber below $a = 0.05$ and 2, respectively.

The discussion starts with the first part of Table 1 where results are given when $C_\eta = 0.0001$. The first set of three columns for $C_\delta = 1$ shows that all the magnitudes of M tend to zero as the wavenumber tends to zero but at different rates. It is shown that the varicose mode is the more unstable for all wavenumbers in the range presented and that the value of the Marangoni number is far smaller than that of the stationary case of the flat wall.

It is interesting to see that, as in our sinuous stationary case of the last section, the second column of this set shows that it is possible to destabilize the fluid layer in the sinuous mode by a relatively small negative Marangoni number. This has no counterpart in the stationary case of $B_\delta = 0$ and therefore it will always be oscillatory.

Another characteristic of the varicose mode of this set is that the value of M changes sign when the wavenumber is between $a = 1$ and 1.1. Therefore, for a positive M the oscillatory varicose mode will be the more unstable when a is smaller than a certain value (here around $a = 1$), but for a larger value stationary convection will be the first to appear. If $M < 0$ the sinuous oscillatory mode M2 will appear but, upon increasing the wavenumber, the oscillatory varicose mode will be the first to occur after a wavenumber between $a = 1$ and 1.1. M2 also changes sign, from negative to positive, at a wavenumber between $a = 1$ and 1.1 by means of an abrupt jump (at a singular a). After that point the positive M2 decreases with a until a minimum value is reached around $a = 1.4$ after which it increases again.

Calculations were made also for $C_\delta = 0.01$. Some similarities in the behavior with the case discussed above were found, but some important differences deserve discussion. Again, the oscillatory varicose mode is the more unstable but not for all wavenumbers. Very near $a = 0.5$ the stationary case for a flat wall becomes the more unstable. In the range of wavenumbers presented the Marangoni number remains positive. However, the second sinuous Marangoni number M2 changes sign at a smaller singular wavenumber between 0.7 and 0.75. The magnitude of M2 is larger than that of $C_\delta = 1$ for all a.

The second set of columns for $C_\delta = 0.001$ shows a behavior similar to that for $C_\delta = 0.01$. Here, the oscillatory varicose mode is the more unstable in a smaller range of wavenumbers which ends between $a = 0.1$ and 0.2. A difference is that the magnitudes of the Marangoni numbers are larger than those of $C_\delta = 0.01$, with the exception of the values M1 and M2 corresponding to $a = 0.01$. In the range of wavenumbers presented, M for the varicose mode changes sign around $a = 1.1$. The value of M2 may change sign after $a = 1.5$ as can be understood due to the large magnitude of the numerical result which approaches to the singular a.

Calculations were made also for $C_\delta = 0.0001$, where the effect of similar large tensions at both the free surface and the membrane is present. The oscillatory varicose mode is the more unstable only until $a = 0.02$, a value after which it was not possible to find solutions for oscillatory convection in a broad range of frequencies investigated. In this way, the sinuous mode M1 is the more unstable from $a > 0.02$ up to a wavenumber between $a = 0.05$ and 0.1. After that value the M for stationary convection on a flat wall is the more unstable. The negative magnitude of M2 is very large even for small a. This might show that, as $a \to 0$, a finite Marangoni number is attained, as was shown under some special conditions in the previous section for the stationary case. It was found that the absolute value of M2 has a maximum and two local minima in the range of a values investigated. However, its overall magnitude is so much larger that the liquid layer may be considered very stable for negative M.

The second part of Table 1 shows results corresponding to a free surface crispation number $C_\eta = 0.01$. Two sets of columns are presented for $C_\delta = 1$ and $C_\delta = 0.001$, but calculations were also made for $C_\delta = 0.01$ and $C_\delta = 0.0001$.

The tenth column shows that here stationary convection is more unstable than in the first part of Table 1 because the tension of the free surface is smaller. The same can be said of the results of the oscillatory convection of the eleventh column, where it is shown that no marginal Marangoni number was found for wavenumbers smaller than $a = 0.3$.

The first set of three columns for $C_\delta = 1$, starting from the twelfth column, shows that the values of M are smaller than those corresponding to the first part of Table 1. The behavior of these Marangoni numbers is similar except for some particular but important points. For wavenumbers from zero to $a < 0.05$ the sinuous mode M1 is the more unstable. After this value the varicose mode will be the first to appear. For negative M the sinuous mode M2 is the more unstable until a is almost 1, after which the varicose mode is the first to appear, that is, for $a > 1$.

The results calculated here for $C_\delta = 0.01$ already present differences with respect to those corresponding to $C_\eta = 0.0001$.

Here it is not clear which mode, the sinuous or the varicose, is the more unstable for very small wavenumbers. However, with increasing a the varicose mode is the first to appear but only for wavenumbers up to $a = 0.181$, which is the last point where M could be calculated. After that the oscillatory sinuous mode becomes the more unstable up to a wavenumber between $a = 0.3$ and 0.4. After this value of a only stationary convection is possible. Another difference found is that here the magnitude of M2 for small wavenumbers is many times larger. It is possible that M2 reaches a finite limit value when $a \to 0$. In the range of wavenumbers calculated the magnitude of M2 does not change much.

The second set of columns of the second part of Table 1 for $C_\delta = 0.001$ presents a behavior similar to the results for $C_\delta = 0.01$ but with an important difference. That is, stationary convection is the one which will appear first. The magnitudes of M in the tenth column are always smaller than those of M1 of the sinuous mode and M of the varicose mode (which is greater than M1). The only way the fluid layer may have oscillatory convection is by means of $M < 0$ as for the sinuous mode M2 in the second column of this set.

In conclusion, for magnitudes of $C_\delta \leq 0.001$ the stationary convection will be the first to appear. This was also verified with the results for $C_\delta = 0.0001$. It is only possible to have oscillatory instability when $C_\delta \leq 0.001$ if a negative Marangoni number is applied. However, as can be seen in Table 1, the magnitude of the negative M2 increases considerably when C_δ decreases, stabilizing the liquid layer. M2 seems to tend to a finite value when $a \to 0$ and has a minimum in the range of a values investigated.

For the conditions of the second part of Table 1 it is concluded that when the tension of the membrane is equal to or less than that of the free surface there is a tendency to favor stationary convection for wavenumbers larger than a particular magnitude. When the membrane tension is larger than that of the free surface stationary convection will always be favored in the range of wavenumbers investigated.

The physical possibility of the appearance of oscillatory convective motion in the case of negative Marangoni numbers is explained in the same way as in the stationary sinuous mode. Relatively small wavenumbers are necessary for the system to be able to neglect viscous and membrane tension effects. In this case, the membrane is allowed to follow easily the perturbations of the free surface deformation. This also explains why for large wavenumbers the preferred instability is the stationary one in the results of Table 1, except those of the varicose mode for $C_\delta = 1$. When $C_\delta = 1$ the M of the varicose mode becomes negative for a relatively large wavenumber. However, this C_δ is so large that it still allows for a sensible membrane deformation at wavenumbers of order 1.

IV. CONCLUSIONS

The thermocapillary instability of a fluid layer with a deformable free surface coating a deformable membrane has been investigated. The sinuous and the varicose modes of instability were taken into consideration. Both expressions of the Marangoni numbers for the stationary and oscillatory convection were calculated analytically. First, numerical calculations of the marginal values of M against a were made for the stationary case. It was shown that when the Biot number of the membrane is zero its deformation plays no role in the instability and that the problem reduces to that of a flat wall. The behavior of M for the varicose mode is very similar to that of the flat wall but it is always smaller. It was shown that the marginal curves for $B_\delta = 1$ are more stable than those for $B_\delta = \infty$ when the wavenumber is smaller than 1, at which the curves for each value of B_δ intersect. Here, the corresponding a is called the intersection wavenumber. This phenomenon, due to the membrane deformation, is in contrast with results already known in the literature on natural and Marangoni convection where an increase in the Biot number of the wall stabilizes the fluid layer. The marginal curves for the sinuous mode also were investigated. The sinuous mode is characterized by negative Marangoni numbers below certain magnitudes of the wavenumber. In fact, those magnitudes of a are singularities of M because they are roots of the denominator of its analytic expression. M changes sign, increasing a above this value. For larger a the magnitudes of M become positive and very similar to those of the varicose mode. The negative Marangoni number brings about the possibility of destabilizing the fluid layer changing the sign of the temperature gradient, which under other conditions should be stabilizing. As in the varicose mode, it was also found that the negative marginal curves for $B_\delta = 1$ are more stable (larger magnitude) than those for $B_\delta = \infty$.

The marginal Marangoni numbers of oscillatory convection present a series of complications due to the variety of possibilities found for instability. Here, due to the large number of nondimensional parameters, only the case in which both Biot numbers are zero is calculated. When the Biot number of the membrane is zero the stationary problem reduces to that described in Ref. (3) of a flat wall. Here, the instability of this problem was calculated numerically again, along with the oscillatory instability related with Ref. (4), for the sake of comparison with the results of the present paper. It has been confirmed that stationary convection appears before the oscillatory one when the wall is flat, as suggested by the graphs of Ref. (4). It has been found that when the tension of the membrane is smaller than or equal to that of the free surface (see Table 1) the varicose mode of oscillatory convection is the first to appear. When the tension of the membrane is increased the range where the varicose mode is more unstable than the stationary case becomes more restricted in the wavenumber range. The oscillatory sinuous mode can be excited first only when the Marangoni numbers are negative. However, the magnitude of M2 increases with the tension of the membrane, making it difficult to destabilize the fluid layer.

When the free surface tension is reduced (second part of Table 1), a weak membrane tension may allow the first oscillatory sinuous mode M1 to be the more unstable in a small range of wavenumbers after which the varicose mode prevails. Increasing the tension of the membrane limits the range in which the varicose mode may exist, allowing the sinuous mode to be the more unstable for a larger range of a. However, above some magnitude of the wavenumber the stationary convection on the flat wall will be the more unstable. When the magnitude of the membrane tension is larger than that of the free surface, stationary convection on a flat wall is the more unstable in the range of wavenumbers shown in Table 1.

It is concluded that it is physically possible to destabilize the fluid layer with negative Marangoni numbers because of the new degrees of freedom added by the membrane deformation. A perturbation given to the free surface by heating from above produces a free surface deformation that is transmitted to the membrane. The perturbation will be opposed by friction at the membrane and by the degree of surface tension and membrane tension. However, depending on the magnitude of the wavenumber this opposition may or may not be important. For relatively small wavenumbers not only viscous but also surface tension and membrane tension effects are not very important and the membrane can follow the motion of the free surface and even may promote the growth of the perturbation. In this way, the perturbation is not dissipated like in a flat wall. It was found that for stationary convection the preferred mode for negative Marangoni numbers is the sinuous mode. For oscillatory convection and fixed heat flux the preferred mode for $M < 0$ is again the sinuous one, but it was found that the varicose mode can also be excited for large membrane crispation numbers.

This variety of possible situations speaks about the richness of phenomena appearing when the membrane (or wall) deformation is included in the problem of Marangoni convection.

ACKNOWLEDGMENTS

The author thanks DGAPA-UNAM for support through project IN119200. Thanks are also due to Mr. Raul Reyes for technical support.

REFERENCES

1. Pearson, J. R. A., *J. Fluid Mech.* **4,** 489 (1958).
2. Vidal, A., and Acrivos, A., *Phys. Fluids* **9,** 615 (1966).
3. Scriven, L. E., and Sternling, C. V., *J. Fluid Mech.* **19,** 321 (1964).
4. Castillo, J. L., and Velarde, M., *J. Fluid Mech.* **125,** 463 (1982).
5. Funada, T., *J. Phys. Soc. Jpn.* **55,** 2191 (1986).
6. Dávalos-Orozco, L. A., *Colloids Surf. A* **157,** 223 (1999).
7. Jain, R. K., Ivanov, I. B., Maldarelli, Ch., and Ruckenstein, E., *in* "Dynamics and Instability of Fluid Interfaces" (T. S. Sorensen, Ed.), Lectures Notes in Physics 105, p. 140, Springer Verlag, New York, 1979.