Three-dimensional convection in a two-layer system with anomalous thermocapillary effect

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At temperatures somewhat above room level, the interfacial tension between a 10 cS silicone oil and ethylene glycol increases with temperature, whereas it typically decreases for other systems of immiscible viscous fluids. The convective flows produced by the combined action of this so-called anomalous thermocapillary effect and buoyancy in this particular liquid–liquid system are studied by direct three-dimensional nonlinear simulation. The liquids are situated between rigid horizontal plates that are kept at different temperatures. A pseudospectral code is used to solve the evolution equations with periodic boundary conditions in the horizontal directions. Depending on the Grashof and Marangoni numbers \( G \) and \( M \), the motionless state can either have a stationary or oscillatory instability. The corresponding finite amplitude solutions show a variety of regular structures such as stationary rolls, stationary hexagons, pulsating hexagons, alternating rolls as well as spatio-temporal chaos. The properties of the alternating rolls are investigated in some detail. Irregular patterns arising from the transition between hexagons and alternating rolls are briefly discussed. © 2002 American Institute of Physics. [DOI: 10.1063/1.1506923]

I. INTRODUCTION

During the last decades, the convective phenomena in the presence of an interface have attracted great attention, specifically due to numerous technological applications.\(^1,2\)

Two main instability mechanisms exist in systems with interfaces. The buoyancy driven (Rayleigh–Bénard) instability appears due to the temperature dependence of the fluid density, and the thermocapillary (Marangoni) instability is generated by the temperature dependence of the interfacial tension: When the temperature grows, the interfacial tension can decrease (the normal thermocapillary effect) or increase (the anomalous thermocapillary effect). The latter type of the temperature dependence of the interfacial tension was observed in aqueous alcohol solutions, nematic liquid crystals, binary metallic alloys, etc. (see, e.g., Ref. 3 and references therein). It also exists in certain liquid–liquid systems such as 10cS silicone oil/ethylene glycol.\(^4\)

Buoyancy (a volume effect) is more important for relatively thick layers, while the thermocapillarity (an interfacial effect) plays the dominant role in the case of thin layers or under microgravity conditions. However, the case where both effects should be taken into account is the most typical. In a one-layer system heated from below, the buoyancy volume forces and the thermocapillary interfacial stresses act in the same direction and produce together a stationary instability, provided the thermocapillary effect is normal.\(^5\) For two-layer systems, the situation is more intricate. If the flow in the lower layer is dominant, the actions of buoyancy and thermocapillary stress are similar to those in the one-layer system. If the flow in the upper layer is dominant, the actions of buoyancy and thermocapillary stress are similar to those in the one-layer system. If the flow in the upper layer is dominant, the buoyancy forces and the thermocapillary stresses act in opposite directions. Their competition leads to a stabilization of the stationary instability, as well as to the generation of a specific kind of linear oscillatory instability, which has been predicted theoretically\(^6\) and observed in the experiments.\(^7\) In the case of the anomalous thermocapillary effect, one can obtain an oscillatory instability when the flow in the lower layer is
dominant, and only a stationary instability in the opposite case.\textsuperscript{4}

It is known that the stationary Marangoni instability generates a hexagonal pattern near the instability threshold.\textsuperscript{5,8,9} This pattern may compete with the square pattern and roll pattern.\textsuperscript{10–14} The buoyancy acts in favor of the roll pattern.\textsuperscript{15,16}

The nonlinear development of the oscillatory instability is much less investigated. In the two-dimensional case, the finite-amplitude regimes of convection that appear due to the oscillatory instability of the mechanical equilibrium were simulated by Braverman et al.\textsuperscript{4} Weakly nonlinear theory in the three-dimensional case faces a serious problem caused by the rotational symmetry of the system. This symmetry leads to a strong degeneracy of the linear eigenvalue problem: The growth rate of a disturbance on the background of a mechanical equilibrium state does not depend on the direction of the wavevector. Near the threshold, the bifurcation analysis can be done by means of the equivariant bifurcation theory only under a certain assumption about the spatial structure of the pattern. Such an analysis has been done for “two-wave” (square-like) patterns\textsuperscript{17–19} and for “three-wave” (hexagon-like) patterns.\textsuperscript{20–22} It is not known \textit{a priori} which of these are relevant for the oscillatory instability in the two-layer problem. Numerical simulations of the full three-dimensional problem provide this information since they do not require any assumptions about the pattern, and are not restricted to small amplitudes.

The present paper reports direct simulations with a pseudospectral code of a 10cS silicone oil–ethylene glycol system under the combined action of buoyancy and anomalous thermocapillary effect (the interfacial tension increases with the temperature). Our work is in line with the recent theoretical interest in the two-fluid approach\textsuperscript{23} as well as with the tendency towards three-dimensional simulations of Bénard–Marangoni convection.\textsuperscript{24} In Sec. II we describe the mathematical formulation of the problem. The main ideas of the numerical method are presented in Sec. III. The results of the nonlinear simulation are given in Sec. IV. Section V contains concluding remarks.

II. FORMULATION OF THE PROBLEM

We consider a layered system of two immiscible liquids with thicknesses $d_1$ and $d_2$, where subscript 1 refers to the top layer and subscript 2 refers to the bottom layer. The interface between the liquids is located at $z=0$, and the upper and lower bounding plates are at $z=d_1$ and at $z=-d_2$, respectively. We disregard the deformation of the interface since it can essentially influence the convective instability only in the case of extremely thin layers.\textsuperscript{25} Between the plates, the temperature difference $\Delta T$ is applied with $T=T_0$ at the top plate and $T=T_0+\Delta T$ at the bottom plate. In the horizontal directions $x$ and $y$ we assume periodic boundary conditions with periodicity lengths $L_x$ and $L_y$.

The relevant material properties of the fluids are the dynamic viscosities $\mu_j$, the kinematic viscosities $v_j$, the densities $\rho_j$, the heat conductivities $\lambda_j$, the thermal diffusivities $\kappa_j$ and the thermal expansion coefficients $\beta_j$, where $j \in \{1,2\}$. The interfacial tension $\sigma$ acting on the fluid interface depends on the temperature according to $\sigma=\sigma_0-\gamma(T-T_0)$ with $\gamma=-0.0127$ mN m$^{-1}$ K$^{-1}$ for the 10cS silicone oil/ethylene glycol system.\textsuperscript{4} We choose $d_1$ as the unit of length, $v_1/d_1$ as the unit of velocity, $d_1^2/v_1$ as the unit of time, $\rho_1 v_1^2/d_1^3$ as the unit of pressure, $T_0$ as reference temperature and $\Delta T$ as temperature unit. In the basic state of pure heat conduction, the temperature depends linearly on the vertical coordinate in each layer. Because of different thermal conductivities, the slope of the temperature profile changes at the interface. The conductive profile is

$$
T_c(z) = \begin{cases}
\frac{d(1-z)}{(\lambda+d)} : & 0 \leq z \leq 1,
\frac{d(1-\lambda z)}{(\lambda+d)} : & -1/d \leq z \leq 0,
\end{cases}
$$

where $f=f_1/f_2$ denotes the ratio of the values $f_1$ and $f_2$ of the corresponding parameter in the layers 1 and 2, respectively, e.g., $d=d_1/d_2$. We can now introduce the temperature perturbation

$$
\theta = T-T_c(z),
$$
as deviation from the distribution in the quiescent state. The velocity fields $v_j=(u_j,v_j,w_j)$ and the temperature perturbations $\theta_j$ in each of the incompressible fluid layers are governed by the Navier–Stokes and energy equations. In Boussinesq approximation, and with our choice of units they take the following dimensionless form:

$$
\vec{\partial}_t v_1 + (v_1 \cdot \nabla) v_1 = -\nabla p_1 + \nabla^2 v_1 + G e_1 \theta_1, \quad (3)
$$

$$
\nabla \cdot v_1 = 0, \quad (4)
$$

$$
P(\vec{\partial}_t \theta_1 + (v_1 \cdot \nabla) \theta_1) = \nabla^2 \theta_1 + \frac{P d}{d+\lambda} w_1, \quad (5)
$$

$$
\nu(\vec{\partial}_t v_2 + (v_2 \cdot \nabla) v_2) = -\mu \nabla p_2 + \nabla^2 v_2 + \frac{G \nu}{\beta} e_2 \theta_2, \quad (6)
$$

$$
\nabla \cdot v_2 = 0, \quad (7)
$$

$$
P \kappa(\vec{\partial}_t \theta_2 + (v_2 \cdot \nabla) \theta_2) = \nabla^2 \theta_2 + \frac{P \kappa d \lambda}{d+\lambda} w_2. \quad (8)
$$

The separation of the linear conductive profile according to Eq. (2) gives rise to the linear terms proportional to $w_j$ in Eqs. (5) and (8). The Grashof number $G$ and the Prandtl number $P$ are defined by

$$
G = \frac{\beta_1 g \Delta T d_1^3}{\nu_1}, \quad P = \frac{\nu_1}{\kappa_1}. \quad (9)
$$

Note that the above evolution equations in both layers can be written in the form

$$
\Lambda_j(\vec{\partial}_t v_j + (v_j \cdot \nabla) v_j) = -\Pi_j \nabla p_j + \nabla^2 v_j + e_j \Gamma_j \theta_j, \quad (10)
$$

$$
\nabla \cdot v_j = 0, \quad (11)
$$

$$
\Sigma_j(\vec{\partial}_t \theta_j + (v_j \cdot \nabla) \theta_j) = \nabla^2 \theta_j + \Omega_j w_j, \quad (12)
$$

where $\Lambda_j$, $\Pi_j$, $\Gamma_j$, $\Sigma_j$, and $\Omega_j$ are dimensionless parameters.

The boundary conditions complementing the evolution equations are
\[ z = 1: \quad \theta_1 = v_1 = 0, \]
\[ z = -1/d: \quad \theta_2 = v_2 = 0 \]

at the top and bottom walls. At the liquid–gas interface \( z = 0 \) the following conditions hold:

\[ \theta_1 = \theta_2, \quad \partial_z \theta_2 = \kappa \partial_z \theta_1, \]
\[ u_1 = u_2, \quad v_1 = v_2, \quad w_1 = w_2 = 0, \]
\[ \mu \partial_z u_1 - \partial_z u_2 = -\frac{\mu M}{P} \partial_z \theta_1, \]
\[ \mu \partial_z v_1 - \partial_z v_2 = -\frac{\mu M}{P} \partial_z \theta_1. \]

Conditions (15) and (16) imply the continuity of temperature and heat flux, conditions (17) describe the continuity of velocity and the nondeformability of the interface, conditions (18) and (19) reflect the balance of the interfacial shear components. The terms on the right hand sides of (18) and (19) are the Marangoni forces. The parameter

\[ M = \frac{\gamma \Delta T d}{\rho_1 v_1 \kappa_1}, \]

is the Marangoni number.

### III. NUMERICAL METHOD

The evolution equations for the hydrodynamic variables are solved using a pseudospectral numerical method.\footnote{26,27} It represents a straightforward extension of the method used in Ref. 28 for one-layer simulations. Apart from the buoyancy terms, it was briefly explained in Ref. 29. We shall present here a brief description which includes the treatment of the buoyancy terms. Notice that the equations are slightly different from Ref. 29 due to the different choice of units.

The discretization in each layer is based on the expansion in Fourier modes with respect to \( x \) and \( y \) and in Chebyshev polynomials in \( z \). In each layer, the same number of Fourier modes is used. Because of incompressibility, only two velocity components are independent. The velocity field in each layer can be represented in terms of two scalar quantities using the poloidal-toroidal decomposition

\[ v_j(x,y,z,t) = \nabla \times (\nabla \times \mathbf{e}_j \Phi_j(x,y,z,t)) + \nabla \times \mathbf{e}_j \Psi_j(x,y,z,t). \]

By that, the incompressibility constraint is satisfied automatically. The evolution equations for \( \Phi_j \) and \( \Psi_j \) are derived by taking the curl and twice the curl of the Navier–Stokes equations and projection onto the vertical direction. We obtain the equations

\[ \Lambda_j(\partial_z \xi_j + N_1(v_j)) = \nabla^2 \xi_j, \]
\[ \Lambda_j(\partial_t \nabla^2 w_j + N_2(v_j)) = \nabla^4 w_j + \Gamma_j \Delta \xi_j, \]

for the vertical velocity component \( w_j = -\Delta \xi \Phi_j \) and the vertical vorticity component \( \xi_j = -\Delta \Psi_j \), where \( \Delta = \partial_x^2 + \partial_y^2 \). \( \Lambda \) denotes the horizontal Laplace operator. The vorticity is defined by \( \mathbf{\omega}_j = \nabla \times \mathbf{v}_j \). The nonlinear terms \( N_1, N_2 \) read

\[ N_1(v) = -\mathbf{e}_z \cdot \nabla \times (\mathbf{v} \times (\nabla \times \mathbf{v})), \]
\[ N_2(v) = \partial_z \nabla \cdot (\mathbf{v} \times (\nabla \times \mathbf{v})) - \mathbf{e}_z \cdot \nabla^2 (\mathbf{v} \times (\nabla \times \mathbf{v})). \]

The velocity components \( u_j \) and \( v_j \) are related to \( w_j \) and \( \xi_j \) by

\[ \Delta \xi u_j = -\partial_z \partial_z w_j - \partial_z \xi_j, \]
\[ \Delta \xi v_j = -\partial_z \partial_z w_j + \partial_z \xi_j, \]

where the continuity equation and the definition \( \xi_j = \partial_z v_j - \partial_z u_j \) have been used. Obviously, \( w_j \) and \( \xi_j \) determine the velocity field up to a mean flow \( U_j(z) \mathbf{e}_x + V_j(z) \mathbf{e}_y \). Equations for \( U_j \) and \( V_j \) are obtained by averaging the Navier–Stokes equations over horizontal cross sections of the periodicity domain. From the Navier–Stokes equations in each layer we find that

\[ \Lambda_j(\partial_t U_j + \partial_z (u_j w_j)) = \partial_z^2 U_j, \]
\[ \Lambda_j(\partial_t V_j + \partial_z (v_j w_j)) = \partial_z^2 V_j, \]

where the angular brackets \( \langle \rangle \) denote horizontal averages over the periodicity domain. The remaining problem in the reformulation of the evolution equations in terms of \( w_j, \xi_j, \theta_j, U_j, \) and \( V_j \) consists in the derivation of appropriate boundary conditions. At the rigid walls \( z = -1/d \) and \( z = 1 \) we have

\[ w_j = \partial_z w_j = \xi_j = U_j = V_j = \theta_j = 0. \]

The condition \( \partial_z w_j = 0 \) is a consequence of the continuity equation and the no-slip condition \( \mathbf{v}_j = 0 \) at the walls. The conditions at the liquid interface \( z = 0 \) become

\[ \theta_1 = \theta_2, \quad \kappa \partial_z \theta_1 = \partial_z \theta_2, \]
\[ w_1 = 0, \quad w_2 = 0, \]
\[ \partial_z w_1 = \partial_z w_2, \quad -\mu \partial_z^2 w_1 + \partial_z^2 w_2 = \frac{\mu M}{P} \Delta \theta_1, \]
\[ \xi_1 = \xi_2, \quad \mu \partial_z \xi_1 = \partial_z \xi_2, \]
\[ U_1 = U_2, \quad \mu \partial_z U_1 = \partial_z U_2, \]
\[ V_1 = V_2, \quad \mu \partial_z V_1 = \partial_z V_2, \]

where the continuity equation has again been used in deriving (33).

The numerical discretization of Eqs. (22), (23), (12) uses a finite-difference method with fixed time step \( \delta t \). A hydrodynamic variable \( \eta \) at time level \( n \delta t \) is therefore represented as

\[ \eta(x,y,z,n \delta t) = \sum_k \eta_k(z) \exp(i k_x x + k_y y), \]

with \( k = (k_x, k_y) \) as wavevector. Here the expansion coefficients \( \eta_k(z) \) are still functions of the continuous variable \( z \).

We now substitute (37) in each of the evolution equations (22), (23), (12) and apply the backward Euler method for the linear terms except the buoyancy term \( \Gamma \Delta \theta_j \) in Eq. (23). This term, as well as all the nonlinear terms are discretized
with the explicit Adams–Bashforth method of the second order. This applies also to the mean flow equations (28) and (29). Using the definitions $\xi = \Delta w$, $D = d/dz$ and $AB\{f\}^{n} = (3^{n} - n^{-1})/2$ from the Adams–Bashforth formula we find the system

$$
(D^2 - k^2 - \frac{\Lambda_j}{\delta t}) \xi^{n+1}_{j,k} = \Lambda_j \left[ \xi^{n}_{j,k} - \frac{\xi^n_{j,k}}{\delta t} + AB\{[N_1(v_j)]_{k}\}^n \right], \tag{38}
$$

$$
(D^2 - k^2 - \frac{\Lambda_j}{\delta t}) \xi^{n+1}_{j,1,k} = \Lambda_j \left[ \xi^{n}_{j,1,k} - \frac{\xi^n_{j,1,k}}{\delta t} + AB\{[N_2(v_j)]_{k}\}^n \right] + AB\{\Gamma, k^2[\theta_{j,k}]^n\}, \tag{39}
$$

$$
(D^2 - k^2)w^{n+1}_{j,k} - \frac{\xi^n_{j,k}}{\delta t} = 0, \tag{40}
$$

$$
(D^2 - k^2)\sum \left( \theta^{n+1}_{j,k} + \Omega_j w^{n+1}_{j,k} \right) = \Sigma_j \left[ \theta^n_{j,k} + AB\{[v_j, \nabla \theta_j]_{k}\}^n \right], \tag{41}
$$

$$
(D^2 - \frac{\Lambda_j}{\delta t}) U^{n+1}_{j} = \Lambda_j \left[ U^{n}_{j} - \frac{U^n_{j}}{\delta t} + AB\{D(u_j w_j)\}^n \right], \tag{42}
$$

$$
(D^2 - \frac{\Lambda_j}{\delta t}) v^{n+1}_{j} = \Lambda_j \left[ v^{n}_{j} - \frac{V^n_{j}}{\delta t} + AB\{D(v_j w_j)\}^n \right], \tag{43}
$$

of equations for the expansion coefficients corresponding to the wavevector $k$ and for the mean flow components at time level $(n+1)\delta t$. Each of the above equations is of the form $(D^2 - \mu)f = g$, i.e., it is a one-dimensional Helmholtz equation. Discretization by means of the Chebyshev-tau method is based on the representation of the function $f$ in the form of the Chebyshev series, and it yields a tri-diagonal system of linear equations for the expansion coefficients of $f$, which can be solved very efficiently. The equations for the lower and upper layers are coupled via the interface boundary conditions. Moreover, the equations for $\xi_j, w_j$, and $\theta_j$ are coupled by the Marangoni boundary condition (33). Because of the coupling we employ a Green’s function technique as in Ref. 30. With the indices $k$ and $n$ omitted for the moment, the solution for the coupled equations (39)–(41) reads

$$
\xi_j = \xi_j^{(p)} + \alpha_j \xi_j^{(1)} + \beta_j \xi_j^{(2)}, \tag{44}
$$

$$
w_j = w_j^{(p)} + \alpha_j w_j^{(1)} + \beta_j w_j^{(2)}, \tag{45}
$$

$$
\theta_j = \theta_j^{(p)} + \alpha_j \theta_j^{(1)} + \beta_j \theta_j^{(2)} + \gamma_j \theta_j^{(3)}. \tag{46}
$$

Functions $\xi_j^{(p)}, w_j^{(p)}$, and $\theta_j^{(p)}$ are particular solutions of the inhomogeneous equations (39)–(41) with zero boundary values of the function itself. The unknown coefficients $\alpha_j, \beta_j$, and $\gamma_j$ multiply the Green’s functions, which satisfy

$$
(D^2 - k^2 - \frac{\Lambda_j}{\delta t}) \xi_j^{(m)} = 0, \tag{47}
$$

$$
\xi_j^{(m)}(0) = 1, \quad \xi_j^{(m)}(z_j) = (-1)^m, \tag{47}
$$

$$
(D^2 - k^2)w_j^{(m)} = \xi_j^{(m)}, \quad w_j^{(m)}(0) = w_j^{(m)}(z_j) = 0, \tag{48}
$$

$$
(D^2 - k^2 - \frac{\Lambda_j}{\delta t}) \theta_j^{(m)} = -\Omega_j w_j^{(m)}, \tag{49}
$$

TABLE I. Ratio of layer depths and material properties used in the simulations.

<table>
<thead>
<tr>
<th>$d$</th>
<th>$\rho$</th>
<th>$\nu$</th>
<th>$\mu$</th>
<th>$\lambda$</th>
<th>$\kappa$</th>
<th>$P$</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>5/9</td>
<td>0.846</td>
<td>0.6493</td>
<td>0.5493</td>
<td>0.6194</td>
<td>1.096</td>
<td>94.0</td>
<td>1.4516</td>
</tr>
</tbody>
</table>

where $m = 1, 2$. Here we have used $z_j$ to denote the position of the rigid wall in the layer $j$, i.e., $z_1 = 1$ and $z_2 = -1/d$. The coefficients $\alpha_1, \alpha_2, \beta_1, \beta_2, \gamma_1, \gamma_2$ are computed by inserting the representation (44)–(46) in the boundary conditions (30)–(33) and solving the resulting $6 \times 6$ system of linear equations. Since the Green’s functions are constant, they need only be computed once at the start of the time integration. We apply the same technique for the solution of the equations for $\xi_j$ and the mean flow components $U_j$ and $V_j$.

The high computational cost of three-dimensional simulations can be met by an efficient parallelization of the algorithm. It is described in Refs. 28 and 31. The essential step in the parallelization concerns the Fourier transform of a three-dimensional array which is distributed across the processors. We use the transposition method. The program has been implemented in C and parallelized using MPI. For the problem sizes considered in this paper, up to 32 processors can be efficiently used on a CRAY T3E computer. Details concerning the parallel performance can be found in Ref. 29.

For the validation of the code the proper computation of both the linear and nonlinear terms has to be verified. First, we set the nonlinear terms equal to zero in the numerical code. This way, the growth or decay rates of eigenmodes can be measured and compared with the exact values. The verification of the nonlinear terms is based on a comparison with nonlinear simulations of low-Prandtl-number Rayleigh–Bénard convection from the literature.

Linearization of Eqs. (22), (23), (12) shows that the vertical vorticity component $\xi$ decouples from the vertical velocity $w$ and $\theta$. We, therefore, need to solve one eigenvalue problem for the growth rates for perturbations of the basic state of pure heat conduction, and one for the decay rates of vertical vorticity modes. The various parameters are set to the values of the 10cS silicone oil–ethylene glycol system listed in Table I.

For $G = 0$, the instability of the basic state is monotonic. For the value $k = 2 \pi/3$ for the dimensionless horizontal wavenumber and a Marangoni number $M = 3643.6$, the exact value of the growth rate is $\alpha_{ex} = 0.0475$. This value is well reproduced with the numerical code. With 17 Chebyshev modes in each layer we obtain $\alpha_{sim} = 0.0476$ for a time step $\delta t = 0.1$ and $\alpha_{sim} = 0.0475$ for a timestep $\delta t = 0.01$. The vertical vorticity eigenmode with $k = 2 \pi/3$ decays exponentially with a decay rate $\alpha_{ex} = -7.549$. Because of this strong decay, smaller timesteps are required to achieve good agree-
ment with our numerical method. The results are $\alpha_{\text{sim}} = -7.28$ for $\delta t = 0.01$ and $\alpha_{\text{sim}} = -7.52$ for $\delta t = 0.001$ (17 Chebyshev modes per layer). Such small timesteps are not necessary in the nonlinear simulations because there is no need to represent strongly damped modes with high accuracy.

For testing the nonlinear and buoyancy terms we use results for the bifurcation from straight rolls to traveling wave convection.\textsuperscript{33} We can adapt the code to this case by turning the fluid interface into a rigid, isothermal wall. Using the same numerical resolution, our data for the measured mean square of the velocity component parallel to the roll axis for the traveling wave case with the Rayleigh number $Ra = 2000$ differ by less than one percent from the value given in Table II of Ref. 33.\textsuperscript{33} We have checked this for both layers.

IV. RESULTS

We will concentrate on the specific oscillatory instability mechanism that appears in two-layer systems due to the competition of buoyancy ($G > 0$) and anomalous thermocapillary effect ($M < 0$).

As shown in Ref. 4, the oscillatory instability arises in the system 10 cS silicone oil–ethylenglycol when the ratio of layers’ thicknesses is $d = 5/9$. The same system of liquids and the same ratio $d$ is studied in the present paper. Table I shows the ratios of the material properties of these two liquids. Besides the oscillatory instability of the purely conductive state, a stationary instability is also possible. Figure 1 (top) shows the stability boundaries (neutral curves) in the $(G,M)$-plane. In region I, the basic motionless state is linearly stable, whereas it is unstable in regions II and III. The type of the instability is determined by the relative “strength” of buoyancy and thermocapillarity. Notice that there is a positive growth rate in both regions II and III, i.e., there will be convective states of finite amplitude on either side of the II-III boundary. For this reason, the results of the linear stability theory (which predicts a transition from stationary to oscillatory instability of the equilibrium state) cannot be directly applied to the transition between stationary and oscillatory finite-amplitude motions.

A. Final states for large aspect ratio

The main objective of the numerical experiments was to determine the possible planforms of convection. Therefore, a series of simulations was started from the same set of random initial conditions of small amplitude. Table II contains the numerical parameters of these runs. In special cases, the final states of previous simulations were used as initial conditions. All simulations listed in Table II were performed with aspect ratios $L_x = L_y = 25$. These relatively large values were chosen to minimize geometric constraints on the patterns. Spatial resolution was $N_x = N_y = 64$ Fourier modes in the horizontal directions and $N_z = 17$ Chebyshev polynomials in the vertical direction in each of the layers. For this resolution, a single time step takes about 1.4 s on four processors of a CRAY T3E computer (300 MHz clock rate), which was the number of processors typically utilized for a single simulation.

During the simulations, integral quantities such as the Nusselt numbers and the mean velocities for both layers were monitored. The runs were stopped either when an inspection of the integral quantities and of interfacial temperature snapshots indicated a converged state (steady or with periodic oscillations), or when no ordered state emerged after a substantial number of time steps. Table II also gives information on the duration of the run.

As usual, the Nusselt numbers are defined as the ratios.
of the total heat flux divided by the conductive heat flux. In the present situation the expressions are
\[ N_1 = \frac{\langle \alpha, \theta_i \rangle - d(\lambda + d)}{\langle \theta_i \rangle - d(\lambda + d)}, \quad N_2 = \frac{\langle \alpha, \theta_i \rangle / d - \lambda(\lambda + \lambda)}{\langle \theta_i \rangle / d - \lambda(\lambda + \lambda)}, \]
where the brackets \( \langle \rangle \) denote averages over the plane \( z = 0 \).

The mean velocities \( V_i \) (\( i \in \{1,2\} \)) are defined as spatial RMS values and measured in units of \( \kappa_i / l_i \) in each layer, i.e., they can be regarded as Péclet numbers. The formal definitions are
\[ V_i = \frac{d_i}{\kappa_i} \left( \frac{1}{L_x L_y} \int_0^{L_x} \int_0^{L_y} d(x, y, z, t) \, dx \, dy \, dz \right)^{1/2}, \]
where the quantities on the right hand side are dimensional. Numerical values for these integral quantities given in Table III for various spatial resolutions and time steps demonstrate that the simulations are sufficiently resolved.

We begin our discussion of the simulation results with the convective regimes generated by the stationary instability (region II in Fig. 1). Generally, two types of stationary patterns have been found. For relatively large values of the Grashof number \( G \), the roll pattern is observed, while for relatively large values of the Marangoni number \( M \), the typical pattern is the hexagonal one (see Fig. 1). Note that this result is in accordance with the prediction of the weakly nonlinear theory and with the results of direct numerical simulations of Rayleigh–Bénard–Marangoni convection in one-layer systems.

Typical roll patterns are shown in Figs. 1(a) and 1(b). One can see that rolls with different orientations can appear in the system. The selection of the orientation is connected with wavenumber selection. In a finite periodic domain the set of allowed wave vectors is discrete, and the possible values of wavenumbers depend on the orientation of rolls. Specifically, the main wavevector of the pattern shown in Fig. 1(a) is \( (k_x = 7k_b, k_y = 0) \) (therefore, \( k = |k| = (k_x^2 + k_y^2)^{1/2} = 7k_b) \), while the main wavevector of the pattern in Fig. 1(b) is \( (k_x = 5k_b, k_y = 3k_b) \) \( (k = 5.8k_b) \), where \( k_b = 2\pi / L_x = 0.251 \) is the basic wavenumber of the computations.

Hexagonal patterns are presented in Figs. 1(c) and 1(d). In the middle of the convective cell the interfacial temperature is lower than that at the boundary between different cells (the dashed lines correspond to negative values of the temperature perturbation \( \theta \)). In contrast to the roll patterns, which were all free of defects, in some of the hexagonal patterns defects persisted even after long integration times. This was observed for \( M = -400 \) and \( M = -500 \) with \( G = 20 \), whereas for \( M = -400 \) and lower \( G \) the pattern was purely hexagonal (cf. Table II). Both the proximity of the II-III boundary and the stronger forcing at higher \( G \) may be responsible for the defects. An example of a pattern with persistent defects is shown in Fig. 1(d). The pattern consists of two coexisting stripes of hexagons and squares with some transition layers between them.

Further increase of the Marangoni number leads to the appearance of the alternating roll pattern (see Fig. 2) which turns out to be the typical kind of oscillatory pattern corresponding to region III in Fig. 1 (cf. Table II). One can see that this kind of pattern is a nonlinear superposition of two systems of standing waves with orthogonal wave vectors. The temporal phase shift between standing waves of different spatial orientations is equal to \( T/4 \), where \( T \) is the full period of oscillations (see the corresponding time series shown in Fig. 3). That is why one observes some kind of roll patterns that change their orientation with the time interval \( T/4 \).

The orthogonal wave vectors of the alternating roll pattern shown in Fig. 2 are \( (k_x = 7k_b, k_y = 0) \) and \( (k_x = 0, k_y = 7k_b) \) since each of the roll patterns appearing during the cycle consist of seven pairs of rolls. However, patterns with the wave vectors parallel to the coordinate axes are typically not attained when starting from random initial conditions. The alternating roll patterns obtained for \( G = 20 \) are oblique (cf. Table II). E.g., for \( M = -2000 \) the wavevector of one of the standing waves has the components \( (k_x = 5k_b, k_y = 0) \).

<table>
<thead>
<tr>
<th>( G )</th>
<th>( M )</th>
<th>( \delta t )</th>
<th>( t_{max} )</th>
<th>Initial cond.</th>
<th>Result</th>
</tr>
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<tbody>
<tr>
<td>12</td>
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<td>0.21</td>
<td>1260</td>
<td>random</td>
<td>rolls</td>
</tr>
<tr>
<td>15.5</td>
<td>-200</td>
<td>0.84</td>
<td>29400</td>
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<td>0.42</td>
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<tr>
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<td>174300</td>
<td>random</td>
<td>alt. rolls, oblique, defects</td>
</tr>
<tr>
<td>15.5</td>
<td>-3000</td>
<td>0.21</td>
<td>56700</td>
<td>random</td>
<td>alt. rolls, oblique, defects</td>
</tr>
<tr>
<td>16</td>
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<td>0.42</td>
<td>39900</td>
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<td>alt. rolls, oblique</td>
</tr>
<tr>
<td>17</td>
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<td>0.63</td>
<td>44100</td>
<td>random</td>
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<tr>
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<td>0.105</td>
<td>9030</td>
<td>random</td>
<td>hexagons</td>
</tr>
<tr>
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<td>-3000</td>
<td>0.105</td>
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<td>31500</td>
<td>random</td>
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</tr>
<tr>
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<td>25200</td>
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</tr>
<tr>
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<td>random</td>
<td>alt. rolls, oblique</td>
</tr>
<tr>
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<td>25200</td>
<td>random</td>
<td>alt. rolls, oblique</td>
</tr>
<tr>
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<td>random</td>
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<tr>
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<tr>
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<td>273000</td>
<td>hexagons</td>
<td>chaotic hex.</td>
</tr>
<tr>
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<td>0.63</td>
<td>462000</td>
<td>hexagons</td>
<td>chaotic hex.</td>
</tr>
</tbody>
</table>
For $M = -2500$ these components are $(k_x = 4k_b, k_y = 6k_b)$, and for $M = -3000$ they are $(k_x = 7k_b, k_y = 2k_b)$. In each of these cases, the wavevector of the second standing wave is perpendicular to that of the first and of equal length. Strict orthogonality of the wavevectors is not always observed. For $G = 20$ and $M = -1000$ the basic wavevectors of the pattern turn out to be $(k_x = 7k_b, k_y = 0)$ and $(k_x = k_b, k_y = 7k_b)$.

As noted earlier for the rolls, the different orientations of the patterns indicate the preference of a certain wave-
number by the system. The patterns for \( G = 20 \) described above suggest that the wavenumber of the constitutive standing waves increases with \( |M| \) because it takes values \( k = \sqrt{50k_b} \) at \( M = -2000 \), \( k = \sqrt{52k_b} \) at \( M = -2500 \) and \( k = \sqrt{53k_b} \) at \( M = -3000 \). However, for \( M = -700 \) the same orientation is observed as for \( M = -3000 \), i.e., \( k \) is initially decreasing with \( |M| \) (for fixed \( G \)). This conclusion should be regarded with some caution since it is based merely on a single simulation at each point \((G,M)\). In any case, the numerical value of \( k \) of about \( 7 k_b = \approx 1.76 \) is in good agreement with linear stability results. Linear theory predicts not only a critical wavenumber \( k_c \approx 1.8 \) for \( |M|/G \approx 50 \), but also that \( k_c \) grows with \( |M|/G \) [cf. Fig. 6(b) of Ref. 4].

The final patterns appear in the simulations only after a transient process which looks chaotic (“preturbulence;” see, e.g., Ref. 34). The duration of this process depends on \( G \) and \( M \) as well as on the initial condition. Since most of the simulations listed in Table II were started from the same set of random initial conditions, the duration of the transient phase can be compared for these runs. The oscillatory instability in region III (Fig. 1) implies an oscillatory transient phase. Figure 4 shows the temporal evolution of the mean velocity of the lower layer for \( G = 20 \) and three different values of \( M \). We see that the transient process becomes longer when \( |M| \) is increased. This behavior is typical of dissipative nonlinear systems subjected to increasingly strong forcing, which eventually leads to the formation of a chaotic attractor. Such an attractor may already be present for \( M = -4000 \) and \( G = 20 \). When this simulation was terminated, it had not evolved into an ordered pattern (cf. Table II). However, power spectra of the \( V_2 \) time series are very similar for this run and for the transient process for \( G = 20 \), \( M = -3000 \).

**B. Global quantities and vertical structure of alternating rolls**

In order to measure the dependence of integral flow quantities of the alternating rolls on \( M \) and \( G \) for fixed wavelength \( k = 7k_b \), we have performed additional simulations in a small domain admitting only a single pair of rolls. For these simulations, \( N_x = N_y = 8 \) Fourier modes and \( N_z = 17 \) Chebyshev polynomials were used in each layer. The periodicity length of this domain is \( L_x = L_y = 25/7 \). Table IV contains data for the oscillation period \( T \), the Nusselt numbers and the mean velocities in each layer. The values given in Table IV are the temporal averages of \( N_i \) and rms temporal averages of \( V_i \) over the oscillation period.

As can be expected, the flow amplitude in each layer grows with \( G \) and \( |M| \). In agreement with the two-dimensional simulations, the flow in the bottom layer is stronger since both the Nusselt and Pécellet numbers are larger in the bottom than in the top layer. The oscillation period \( T \) decreases as \( |M| \) grows (for fixed \( G \)). This observation again coincides with the predictions of linear stability for the critical mode, i.e., the nonlinear correction to the period does not change the dependence \( T(|M|) \) qualitatively. The period of the critical mode is a decreasing function of \( |M|/G \).[4]

Naturally, a finite flow amplitude causes anharmonic oscillations. This effect is obvious from the two phase diagrams of Fig. 5, which are constructed from the two major Fourier amplitudes \( A_1 \) and \( A_2 \) explained in the caption of Fig. 3. Near the I-III boundary \((G = 15.5, M = -2000)\) the flow amplitude is weak and the phase diagram [Fig. 5(a)] almost circular. It is significantly deformed for \( G = 20 \) and \( M = -3000 \) [Fig. 5(b)].

The accuracy of our numerical data has been checked by performing simulations with higher resolution and smaller timestep for two sets of parameters. The integral velocities \( V_i \) reported in Table IV changed by about 4% for \( G = 15.5, M = -1000 \) and by about 1% for \( G = 20, M = -3000 \) when the numerical parameters were changed to \( \delta t = 2.1 \times 10^{-2} \), \( N_x = N_y = 16 \), and \( N_z = 33 \). Even smaller are the changes in the oscillation period \( T \): about 1% for \( G = 15.5, M = -1000 \) and less than 0.1% for \( G = 20, M = -3000 \). These results indicate that the large aspect ratio computations summarized in Table II are sufficiently resolved as well. We also confirmed that the vertical vorticity is essentially negligible. For \( G = 20, M = -3000 \) its dimensionless rms value in each layer (measured in units of \( \kappa / \delta_i^2 \)) is about two orders of magnitude smaller than the mean velocity.

As another interesting issue we shall now examine the kinetic energy balance of alternating roll convection. In each layer, energy is produced by buoyancy and dissipated through viscosity. The work done by the Marangoni forces represents an additional production term, and it is of interest to see how this energy is distributed to each of the layers.
The kinetic energy balance of each layer (in dimensional units) is obtained by multiplying the Navier–Stokes equation with the velocity and integrating over the volume $V_i$ of the layer. The result is
\[
\frac{d}{dt} \int_{\Omega_i} \frac{\rho V^2}{2} d\Omega = \int_{\Omega_i} \mathbf{v}_i \cdot \mathbf{T}_i d\mathbf{x} dy + \int_{\Omega_i} \rho_i \beta \nabla T_i \cdot \mathbf{w} d\Omega + \oint_{\Gamma_i} \rho_i v_i (\mathbf{n} \times \mathbf{v}_i) \cdot d\mathbf{n},
\]
where $\mathbf{n}_i$ denotes the outer normal vector of layer $i$ on the interface $z=0$, $\mathbf{T}_i$ stands for the viscous stress tensor, and the energy, buoyancy and dissipation terms have been named $E_i$, $B_i$, and $D_i$, respectively. We can now add these equations for layers one and two. Upon using the interfacial boundary conditions we find that
\[
\frac{d}{dt}(E_1 + E_2) = -\int_{z=0} \gamma V \nabla T d\mathbf{x} dy + (B_1 + B_2) - (D_1 + D_2) - P_s,
\]
where $P_s$ is the energy production due to the Marangoni effect. Upon taking the temporal average (indicated by overbar), this equation becomes
\[
\bar{D}_1 + \bar{D}_2 = \bar{P}_s + \bar{B}_1 + \bar{B}_2.
\]
In the simulations summarized in Table IV, this relation is satisfied at least to within 0.1%. Table V shows the total average dissipation $\bar{D}_1 + \bar{D}_2$ in units of $V \rho L_x L_y / d$ as well as $\bar{B}_1$, $\bar{B}_2$, $\bar{D}_1$ and $\bar{D}_2$ normalized to $\bar{P}_s$. The last three columns show the fractions of $\bar{P}_s$ dissipated in layers 1 and 2 given by $R_i = (\bar{D}_i - \bar{B}_i) / \bar{P}_s$ and the ratio $R_2 / R_1$. We see that the stronger convection in the lower layer is associated with a higher total energy dissipation, and that it also receives the larger share of the thermocapillary energy input $\bar{P}_s$. Not surprisingly, the relative magnitude of this production term increases with the absolute value of the Marangoni number. However, the discrepancy in the distribution of this energy input between both layers is more pronounced for lower Marangoni numbers.

To complete our examination of the alternating roll solutions we shall now consider their three-dimensional spatial structure. The alternating rolls are characterized by oscillations.
tions in the Fourier amplitudes corresponding to orthogonal horizontal wavevectors. We have observed that this leads to the appearance of one-dimensional temperature distributions at the interface between the layers during the oscillation period, which is also typical for stationary roll convection. One might therefore ask if the flow field also comes close to a roll state during the oscillation. To answer this question we consider the spatial mean square values $V_x^{ms}$ and $V_y^{ms}$ of the horizontal velocity components $u$ and $v$ in each layer. Figure 6 shows the temporal evolution of these quantities for a single oscillation period for parameters $G = 15.5$ and $M = -1000$. We see that in the top layer, the mean square horizontal velocities become nearly zero, i.e. the flow becomes approximately two-dimensional. This is in contrast with the bottom layer, where the oscillation amplitude is small compared to the mean value. The flow in the bottom layer is therefore always three-dimensional. Isotherms and velocity vectors in the $(x,z)$-plane plotted in Fig. 7 illustrate these different flow structures. Plot (a) corresponds to a minimum of the mean square velocity $V_y^{ms}$ in the top layer, and plot (b) to a minimum of the mean square velocity $V_x^{ms}$.

Similar behavior is found for the other parameter values studied in Table IV.

The structure of the velocity field for the alternating rolls can be roughly approximated by formulas

$$u \sim U(z) \cos(kx) \cos(\omega t - \phi(z)),$$
$$v \sim U(z) \cos(ky) \cos(\omega t - \phi(z) + \pi/2).$$

These expressions can be obtained near the linear instability threshold in the leading order of an asymptotic expansion. The phase $\phi(z)$ appears because the velocity component of the eigenfunction, which is determined by the linear stability theory, is a complex function in the case of an oscillatory instability. We can conclude that the phase $\phi(z)$ is almost constant in the top layer, but it is not so in the bottom layer. The dependence of $\phi$ on $z$ causes persistence of a three-dimensional flow in the bottom layer.

The same analysis may be applied to the temperature perturbation $\theta$. Figure 6(b) shows the temporal evolution of the spatial mean square values $T_x^{ms}$ and $T_y^{ms}$ of $\partial_x \theta$ and $\partial_y \theta$. We can see that the temperature field becomes nearly two-

![Fig. 6. Alternating rolls with $L_x = L_y = 25/7$, $G = 15.5$ and $M = -1000$: (a) Temporal evolution of the spatial mean square values $V_x^{ms}$ and $V_y^{ms}$ (dashed) for the top layer (large amplitude) and the bottom layer (small amplitude). The quantities are normalized in the same way as the Péclet numbers in Eq. (52). (b) Temporal evolution of spatial mean square values $T_x^{ms}$ and $T_y^{ms}$ (dashed) in the top layer (large amplitude) and bottom layer (small amplitude).](image)

![Fig. 7. Snapshots of the velocity field and isotherms in the $(x,z)$-plane for alternating rolls with $L_x = L_y = 25/7$, $G = 15.5$ and $M = -1000$ for approximately two-dimensional flow in the top layer. (a) Top layer flow in the plane and (b) top layer flow perpendicular to the plane.](image)
dimensional in both top and bottom layer, although at different times. This means that the corresponding phase $\tilde{\phi}(z)$ for $\theta$ is nearly constant in each layer.

Note that the same conclusions concerning $\phi(z)$ and $\tilde{\phi}$ can be obtained from the results of 2D nonlinear simulations.\textsuperscript{4} Figure 10 of this paper shows that the distortions of isotherms evolve more or less synchronously in each layer, and that the stream function in the top layer has a simple one-vortex structure. The sign of the vortex is changed almost simultaneously over the whole top layer. On the other hand, the motion in the bottom layer has sometimes a more complicated, two-storey structure. That means that the dependence $\phi(z)$ in the expression (56) for $u$ is significant.

C. Finite amplitude convection in the transition region

The region near the II-III boundary in Fig. 1 has been investigated by means of additional simulations for $L_x = L_y = 25$ with different initial conditions. Let us discuss in more detail the transition between steady hexagons to alternating rolls for the fixed value $G = 20$ when $|M|$ grows.

For $M = -500$, the alternating rolls are unstable, and the stable pattern is the hexagonal one. The hexagons are obtained if the simulation is started with alternating rolls (obtained at a higher value of $|M|$) as the initial condition. The hexagons are obtained also from random initial condition (with defects, see Table II). For $M = -550$, the steady hexagonal pattern becomes unstable. Starting from stationary hexagons (obtained at a lower value of $|M|$), we observe periodic pulsations of the hexagonal pattern (see Fig. 8). The appearance of pulsating hexagons may be caused by a secondary instability of steady hexagons studied in Ref. 22 by means of amplitude equations. The hexagonal structure is conserved for this solution, but some periodic changes of the isotherms’ shapes are evident. They are caused by sinusoidal oscillations of the amplitudes of the wavevectors ($k_x = 5k_b, k_y = \pm 6k_b$). In addition to the oscillation of these modes, the amplitudes of the basic modes of the hexagonal pattern also oscillate slightly about a nonzero average. The stability region of regularly oscillating hexagons overlaps with the stability region of chaotically oscillating hexagons. The latter kind of patterns has been obtained for the same value $M = -550$ as the former one but starting from random initial conditions. The irregular hexagonal patterns persist at $M = -570$, for which Fig. 9(a) shows a snapshot of the interfacial temperature and Fig. 9(b) shows the chaotic evolution of a Fourier amplitude.

For $M = -600$, the hexagonal pattern is destroyed. Starting with hexagons as the initial condition, we have obtained the transition to stable alternating rolls. At the same time, there exists another stable spatial structure at the same value

\begin{figure}[h]
\centering
\includegraphics[width=\columnwidth]{fig8.png}
\caption{Snapshots of the interfacial temperature field during one period of oscillations for $G=20$, $M = -550$ (oscillating hexagons).}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\columnwidth]{fig9.png}
\caption{(a) Snapshot of the interfacial temperature for chaotic hexagons. (b) Time series of the real part of the Fourier amplitude of the interfacial temperature field with wavevector ($k_x = 0, k_y = 7k_b$). The parameters are $G = 20, M = -570$.}
\end{figure}
of $M$. Figure 10 shows the final state attained in a simulation for $M = -600$, which was started from random initial conditions. In this case one observes a competition, rather than a coexistence, of rolls with different orientation, which leads to their spatial separation. The pattern consists of two domains of rolls with different orientation separated by domain walls which look like strips of squares. The square cells are "absorbed" on one end of the roll and "emitted" on another one, thus the domain walls move as a whole. Though that is not clearly seen in Fig. 10, we observe also some oscillations of domain walls. Notice that the kinetic energy of each layer is constant for this case.

Thus, we have found that the stability regions of two generic patterns, steady hexagons and strictly periodic alternating rolls, are separated by two regions where the patterns reveal multistability and less generic behavior. A region of nonstationary hexagonal structures is adjacent to the of steady hexagons. Also, in a certain region of parameters the stable alternating rolls coexist with another pattern where rolls of different orientation are spatially separated by domain walls.

V. SUMMARY AND CONCLUSIONS

We have investigated three-dimensional nonlinear convective patterns in a fluid system with an interface under the combined action of buoyancy and anomalous thermocapillary effect. In the presence of two types of primary instabilities of the mechanical equilibrium state (stationary instability and oscillatory instability), a variety of convective regimes (stationary rolls, stationary hexagons, periodically and chaotically pulsating hexagons, alternating rolls and spatiotemporal chaos) have been found by means of direct three-dimensional simulations with a pseudospectral Fourier–Chebyshev code.

In the region of oscillatory instability, the most typical flow regime turned out to be the alternating roll pattern. Note that this pattern is one of the generic wave patterns that appear in the rotationally invariant systems due to a primary oscillatory instability of the spatially homogeneous state. This prediction from the general theory of dissipative wave patterns can be found in several references including Refs. 17–19. Alternating rolls have been obtained previously in the problem of compressible magnetocoevolution. Clune and Knobloch \cite{35} obtained them from a weakly nonlinear analysis, and Matthews, Proctor, and Weiss \cite{36} observed them in direct numerical simulations. Alternating rolls were observed also in simulations of the Maxwell–Bloch laser equations. \cite{37}

We have studied the properties of alternating roll patterns in detail since this particular pattern is new in the context of Rayleigh–Bénard–Marangoni convection. In particular, we have measured certain integral quantities (cf. Tables IV and V) and determined the vertical structure of the flow. We also obtained some information on wavenumber selection. We hope that these predictions serve as an incentive to study the system also experimentally.

Finally, we discussed some simulations devoted to the transition between finite-amplitude patterns generated by the stationary instability and those generated by the oscillatory instability. The nature of this transition is a rather nontrivial question. In low-dimensional dynamical systems, such a transition takes place typically through a global (homoclinic) bifurcation. In our case, this transition is much more complicated and includes the appearance of chaotic regimes and regimes with spatial defects.

ACKNOWLEDGMENTS

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