conductivity of the circuit; τ, oscillation period; m, volume per fraction of the second component in unit volume of the medium.

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THEORY OF INTERFACIAL CONVECTION

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UDC 532.7:536.25

Using a simple thermocapillary convection problem as an example, we consider weakly nonlinear convective structures which arise near the surface of two liquids as a result of the Marangoni instability.

In the so-called Marangoni instability (a brief historical review is in [1]), near the surface between two immiscible liquids interesting stationary motion can develop. In some cases the motion is characterized by a high degree of order and there is a completely regular circulatory flow inside separate convection rolling cells which form a coherent structure (interfacial convection). In other cases the motion resembles random fluctuations in a turbulent fluid (interfacial turbulence). The different types of motion near the surface were observed in [2-6]; they can also be considered as dissipative structures developing in a nonequilibrium system [7, 8] or as a consequence of the randomizing behavior of dynamical systems with strange attractors [8, 9].

The presence of interfacial convection or turbulence leads to a significant increase in mass or heat transport across the surface (see [10, 11]) and is therefore of great interest. Physically, the Marangoni instability is most often due to the dependence of the surface tension on temperature or concentration of surface-active or inactive material (thermocapillary or concentration-capillary convection), but may also be caused by a dependence of the surface tension on the density of surface electric charges or dipoles, the polarization of the surface layer in an external electromagnetic field, the conformational structure of the surface layer, and so on (see [12]).

The theoretical studies in this field are almost entirely devoted to the linear analysis of the conditions for the onset of the Marangoni instability (representative examples can be found in [13-19]). Attempts to extend the analysis to nonlinear effects are rare [20-22], and except for numerical investigations, are limited to weakly nonlinear problems of interfacial convection. The method used in the present paper is essentially a variant of the small parameter method, applied previously to natural thermal convection in [23] and considered in detail in [24]. It is quite similar to the method of Lin [22, 25] and is based on the old classical works of Stuart and Watson [26, 27] on the nonlinear stability of plane Poiseuille and Couette flow. A similar method was applied to nonlinear instabilities and to the formation of space–time structures in thin liquid films deposited on substrates [28-30].

In essence, the method goes back to the well-known hypotheses of Landau [31] and Hopf [32] that the transition to turbulent motion can be thought of as a series of supercritical bifurcations of the set of periodic (or quasiperiodic) solutions of the Navier–Stokes equations describing the loss of stability in the analogous set of higher dimensionality, and on the possibility of stabilizing these solutions for not very large supercriticalities due to nonlinear interactions (the possibility of establishing regular periodic secondary flow under certain conditions). Although prior to the onset of natural turbulence, this hypothesis was shown to be untrue [33], it is correct in many other cases, in particular for interfacial convection.

A. M. Gor'kii Urals State University, Sverdlovsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 48, No. 2, pp. 230-239, February, 1985. Original article submitted November 23, 1983.

0022-0841/85/4802-0163$09.50

Below our method is illustrated on a plane thermocapillary convection problem. In this case the nonlinearities in the Navier–Stokes equation and heat-conduction equation and also nonlinearities from distortion of the surface and the dependence of the surface tension on the temperature of the surface can be significant in stabilizing finite perturbations. The analysis of the dynamics of nonlinear perturbations leads to very lengthy and complicated calculations such that the basic ideas of the method and the physics of the problem can easily be hidden in a mass of details. In order to avoid this, we consider only the simplest situation, when the only important nonlinearity in the problem is the dependence of the surface tension on temperature. This corresponds to the assumption that the Reynolds and Peclet numbers, characterizing steady convective motion and its effect on heat transport, are small, and the surface is practically undeformable (the surface tension is large). We simplify the problem further by ignoring the temperature dependence of the densities, specific heats, viscosities, and thermal conductivities of both liquids, and consider only plane flow. Then in both liquids we can introduce the stream function (such that $v_x=\partial\psi/\partial z$, $v_z=-\partial\psi/\partial x$), and completely ignore the usual Taylor instability of the surface in an external gravitational field.

The unperturbed state is assumed to be a double-layer of two immiscible liquids at rest, occupying the half-spaces $z < 0$ and $z > 0$ (regions 1 and 2, respectively), in which there is heat transport normal to the plane of the surface ($z = 0$) so that we have a nonequilibrium system. The unperturbed temperature field is given by

$$\tau_0^{(j)} = \tau_{s0} - \beta_j z, \quad j = 1, 2, \tag{1}$$

and from the condition that the heat flux be continuous, it follows that

$$\lambda_1\beta_1 = \lambda_2\beta_2. \tag{2}$$

The linearized equations for the stream function in the two regions, corresponding to the approximation of overdamped motion, and the appropriate boundary conditions have the form

$$\begin{gathered}(\nu\Delta - \partial/\partial t)\,\Delta\psi = 0, \quad \psi\to 0, \quad z\to\pm\infty,\\ \frac{\partial\psi_1}{\partial x} = \frac{\partial\psi_2}{\partial x} = 0, \quad \frac{\partial\psi_1}{\partial z} = \frac{\partial\psi_2}{\partial z}, \quad z=0,\\ \sigma_{xz}^{(2)} - \sigma_{xz}^{(1)} = \mu_2\left(\frac{\partial^2\psi_2}{\partial z^2} - \frac{\partial^2\psi_2}{\partial x^2}\right) - \mu_1\left(\frac{\partial^2\psi_1}{\partial z^2} - \frac{\partial^2\psi_1}{\partial x^2}\right) = -\frac{\partial\sigma}{\partial x}, \; z=0,\end{gathered} \tag{3}$$

where the last equation describes the balance of the tangential components of momentum on the surface (where there is no danger of misunderstanding, the subscripts $j = 1, 2$ denoting liquids 1 and 2, are omitted).

The equations of heat conduction for a temperature perturbation τ, linearized for the case of small Peclet number, and the appropriate boundary conditions have the form

$$\begin{gathered}\partial\tau/\partial t - \beta v_z = \varkappa\Delta\tau, \quad \tau\to 0, \quad z\to\pm\infty,\\ \tau_1 = \tau_2, \quad q_1 = -\lambda_1\frac{\partial\tau_1}{\partial z} = q_2 = -\lambda_2\frac{\partial\tau_2}{\partial z}, \quad z=0.\end{gathered} \tag{4}$$

Equations (4) will be valid in the case where the temperature gradients β_j are large, so that in each half-space the quantity $|\beta v_z|$ is much larger than $|v\nabla\tau|$, which is assumed small. It is not difficult to show that this implies that the following inequality is satisfied:

$$\beta L \gg \tau_s^0 \tag{5}$$

in both regions, where L and τ_s^0 are the linear dimension of the steady convection cells and the characteristic temperature perturbation on the surface, respectively.

We assume a nonlinear relation between the surface tension and surface temperature in the form

$$\sigma(\tau_{s0} + \tau_s) = \sigma - a\tau_s + b\tau_s^2 + c\tau_s^3 + \ldots, \quad \sigma = \sigma(\tau_{s0}), \quad \tau_s = \tau|_{z=0}, \tag{6}$$

and the coefficients a, b, c, ... can be easily expressed in terms of derivatives of σ with respect to τ_{s0}, and are considered to be known.

We consider below only weakly nonlinear periodic convective flow. The periodic functions describing the flow will be nearly harmonic. We therefore assume that these periodic functions can be written is the sum of the principal harmonic and a series of higher harmonics with coefficients that decrease with the degree of the harmonic. For example, the stream function is written as

$$\psi = \varepsilon^2\Psi_0 + \sum_{n=1}^{\infty} \varepsilon^n (\Psi_n e^{in\theta} + \Psi_n^* e^{-in\theta}),$$
$$\theta = \varepsilon^2\omega t - kx, \quad \Psi_1 = \Psi_{10} + \varepsilon^2\Psi_{12} + \dots, \tag{7}$$

where Ψ_n are functions of z only, the wavenumber k is real, and the parameter ε is introduced only to help identify terms of various orders in the amplitude of the principal harmonic; in the final results ε will be set equal to unity. We simplify the calculation further by assuming in (7) that ω is small quantity of the order of the amplitude of the principal harmonic squared. Although the loss of stability in systems of this kind usually occurs with respect to zero-frequency perturbations, the frequency of stationary space–time structures established as a result of the instability is not always small. Therefore, the condition that the complex frequency be small must be verified a posteriori. The representation (7) can also be written for the other variables; the coefficients in the series will be denoted by corresponding capital latters. Below we will consider only the harmonics n = 0, 1, 2.

Substitution of (7) into (3) leads to the equations

$$\Delta_1^2\Psi_{10} = 0, \quad d^4\Psi_0/dz^4 = 0, \quad \Delta_2^2\Psi_2 = 0,$$
$$\Delta_1^2\Psi_{12} = i(\omega/\nu)\Delta_1\Psi_{10}, \quad \Delta_n = d^2/dz^2 - n^2k^2 \tag{8}$$

with boundary conditions following from (3). Using the condition at infinity and the vanishing of the normal component of the velocity on the surface, the coefficients in (7) for the velocity components v_x and v_z take the following form in the half-space $z > 0$:

$$V_{0x} = V_{0z} = 0, \quad V_{10x} = B(1-kz)e^{-kz}, \quad V_{10z} = ikBze^{-kz},$$
$$V_{2x} = B'(1-2kz)e^{-2kz}, \quad V_{2z} = 2ikB'ze^{-2kz},$$
$$V_{12x} = -\frac{i\omega}{4k\nu}Bz(2-kz)e^{-kz}, \quad V_{12z} = \frac{\omega}{4\nu}Bz^2e^{-kz}, \tag{9}$$

where B and B' are constants of integration. The arbitrary constant appearing when the equation for Ψ_{12} in (8) is integrated is chosen so that the term proportional to z vanishes in the expression for Ψ_{12}.

The coefficients of the temperature perturbation in the form (7) follow from (4):

$$\varkappa\Delta_1 T_{10} = -\beta V_{10z}, \quad d^2T_0/dz^2 = 0,$$
$$\varkappa\Delta_2 T_2 = -\beta V_{2z}, \quad \varkappa\Delta_1 T_{12} = -\beta V_{12z} + i\omega T_{10}, \tag{10}$$

and the boundary conditions have the same form as in (4). Solving (10) in the half-space $z > 0$ and using the condition at infinity, we have

$$T_0 = 0, \quad T_{10} = Ae^{-kz} + \frac{i}{4k}\frac{\beta}{\varkappa}Bz(1+kz)e^{-kz},$$
$$T_2 = A'e^{-2kz} + \frac{i}{8k}\frac{\beta}{\varkappa}B'z(1+2kz)e^{-2kz},$$
$$T_{12} = -\frac{i\omega}{2k\varkappa}Aze^{-kz} + \frac{\omega\beta}{8k^3\varkappa}Bz\left[\left(\frac{1}{\varkappa}+\frac{1}{2\nu}\right)(1+kz) + \frac{k^2z^2}{3\varkappa}\right]e^{-kz}, \tag{11}$$

where A and A' are new constants of integration. In the solution of the equation for T_{12} in (10), the arbitrary constant is chosen to cancel terms not involving powers of z.

Relations for the half-space $z < 0$ (region 1) corresponding to (9) and (11) can be obtained from the above equations by simultaneously changing the sign of i, k, and ω.

Below we will need the values of the coefficients $U_n = V_{nx}$, Σ_n, T_{sn}, and Q_n for the tangential velocity and stress, temperature, and heat flux at the surface in both directions. We obtain from (9)

$$U_0^{(j)} = \Sigma_0^{(j)} = 0, \quad U_{10}^{(j)} = B_j, \quad U_2^{(j)} = B_j', \quad U_{12}^{(j)} = 0,$$
$$\Sigma_{10}^{(j)} = (-1)^{j-1} 2k\mu_j B_j, \quad \Sigma_2^{(j)} = (-1)^{j-1} 4k\mu_j B_j', \tag{12}$$
$$\Sigma_{12}^{(j)} = (-1)^{j-1} (i\omega\rho_j/2k) B_j, \quad j = 1,\ 2,$$

and from (11)

$$T_{s0}^{(j)} = Q_0^{(j)} = 0, \quad T_{s10}^{(j)} = A_j, \quad T_{s2}^{(j)} = A_j', \quad T_{s12}^{(j)} = 0,$$
$$Q_{10}^{(j)} = (-1)^j k\lambda_j A_j - \frac{i\rho_j C_j \beta_j}{4k} B_j,$$
$$Q_2^{(j)} = (-1)^j 2k\lambda_j A_j' - \frac{i\rho_j C_j \beta_j}{8k} B_j', \tag{13}$$
$$Q_{12}^{(j)} = \frac{\omega\rho_j C_j}{2k}\left[(-1)^j iA_j - \frac{\beta_j}{4k^2}\left(\frac{1}{\varkappa_j} + \frac{1}{2\nu_j}\right) B_j\right], \quad j = 1,\ 2.$$

In (12) and (13) the subscripts indicate the region to which the expression refers.

The unknown constants A_j, B_j and A_j', B_j' are determined from the remaining boundary conditions in (3) and (4). We use the Fourier expansion (7) for all quantities in the boundary conditions and write separate relations for each harmonic. The continuity conditions of the tangential components of the velocity and the temperature give

$$A_1 = A_2 = A, \quad B_1 = B_2 = B, \quad A_1' = A_2' = A', \quad B_1' = B_2' = B'. \tag{14}$$

From the balance of the tangential components of the momentum and the heat flux on the surface in (3) and (4), we obtain the following relations with the help of (6) and (12) through (14). For the first harmonic we have

$$\left[2(\mu_1 + \mu_2) + \varepsilon^2 \frac{i\omega}{4k^2}(\rho_1 + \rho_2)\right] B = i\{aA - \varepsilon^2 [b(AA'^* + A^*A') + 3cA^2A^*]\},$$
$$-\frac{i}{4k^2}\left\{\rho_1 C_1 \beta_1 - \rho_2 C_2 \beta_2 - \varepsilon^2 \frac{i\omega}{2k^2}\left[\rho_1 C_1 \beta_1\left(\frac{1}{\varkappa_1} + \frac{1}{2\nu_1}\right) - \right.\right. \tag{15}$$
$$\left.\left. - \rho_2 C_2 \beta_2\left(\frac{1}{\varkappa_2} + \frac{1}{2\nu_2}\right)\right]\right\} B = \left[\lambda_1 + \lambda_2 + \varepsilon^2 \frac{i\omega}{2k^2}(\rho_1 C_1 + \rho_2 C_2)\right] A,$$

and for the second

$$2(\mu_1 + \mu_2) B' = aA' - bA^2,$$
$$-\frac{i}{8k^2}(\rho_1 C_1 \beta_1 - \rho_2 C_2 \beta_2) B' = 2(\lambda_1 + \lambda_2) A'. \tag{16}$$

The system of equations (16) determines A' and B' as functions of A. In particular

$$A' = \frac{br(ar + 4isk^2)}{a^2r^2 + 16s^2k^4} A^2, \tag{17}$$

where

$$r = \rho_1 C_1 \beta_1 - \rho_2 C_2 \beta_2; \quad s = 8(\lambda_1 + \lambda_2)(\mu_1 + \mu_2). \tag{18}$$

Substituting (17) into (15), we obtain a system of linear, homogeneous equations for A and B with coefficients dependent on the amplitude squared $\delta = AA^*$. The characteristic equation of the system (to order ε^2) has the form

$$\varepsilon^2 i\omega\left(\frac{R}{k^2}+S\right)=ar-k^2s-\varepsilon^2 r\left[\frac{2b^2r\,(ar+4isk^2)}{a^2r^2+16s^2k^4}+3c\right]\delta, \tag{19}$$

where

$$R=\frac{a}{2}\left[\rho_1C_1\beta_1\left(\frac{1}{\varkappa_1}+\frac{1}{2\nu_1}\right)-\rho_2C_2\beta_2\left(\frac{1}{\varkappa_2}+\frac{1}{2\nu_2}\right)\right];$$
$$S=(\rho_1+\rho_2)(\lambda_1+\lambda_2)+4(\rho_1C_1+\rho_2C_2)(\mu_1+\mu_2). \tag{20}$$

Letting $\omega = -\Omega - i\gamma$, where Ω and γ are real, and putting $\varepsilon = 1$, we obtain from (19)

$$\gamma=\gamma'(k)+\gamma''(k)\,\delta,\quad \Omega=\Omega''(k)\,\delta,$$
$$\gamma'(k)=k^2\frac{ar-sk^2}{R+Sk^2},\quad \gamma''(k)=-\frac{rk^2}{R+Sk^2}\left[\frac{2ab^2r^2}{a^2r^2+16s^2k^4}+3c\right],$$
$$\Omega''(k)=\frac{8b^2r^2sk^4}{(R+Sk^2)(a^2r^2+16\,s^2k^4)}, \tag{21}$$

where r, s, R, S are given in (18) and (20).

The linear stability theory corresponds to $\delta = 0$ in (19) and (21). In particular, when $R + Sk^2 > 0$, the states of neutral stability are determined by the equation $ar = sk^2$, and since we always have $s > 0$ and usually have $a > 0$, we must have $r > 0$ in order to have an instability. (When $R < 0$, the equation $R + Sk^2 = 0$ also has a positive root, but since we assumed that ω was small, we cannot consider values of k close to this root using the relations obtained here.)

If the unperturbed state is unstable, and stationary nearly harmonic convective flow is established with characteristic linear dimension k_*^{-1} and amplitude $\delta_*^{1/2}$, the growth factor of the perturbation γ must go to zero when $k = k_*$ and $\delta = \delta_*$. Also this zero value must correspond to the maximum value of γ, considered as a function of k at δ = const (for a detailed discussion of this, see [29, 30]). Hence we obtain from (21) the following system of equations for the unknown k_* and δ_*:

$$\gamma'(k_*)+\gamma''(k_*)\,\delta_*=0,\qquad \frac{\partial\gamma'(k_*)}{\partial k_*}+\frac{\partial\gamma''(k_*)}{\partial k_*}\delta_*=0, \tag{22}$$

and the value of γ corresponding to the roots k_* and δ_* of this system must be a maximum, in the sense discussed above.

If the system (22) has positive roots which satisfy the maximum condition on γ, then the loss of stability is "soft." In this case, an ordered, stationary, periodic interfacial convection regime can be established near the surface with a definite frequency and with convective cells of a definite linear dimension. This regime is evidently stable against small plane perturbations with arbitrary k and Ω. Such perturbations can be considered as weak modulations imposed externally on the unperturbed system [24], whose amplitude is too small to change the stability characteristics of the system. (However, this regime is not always stable against spatial perturbations; for certain values of the parameters it can be replaced by a stable spatial regime with a coherent structure of three-dimensional cells, rather than two-dimensional ones. A similar situation is observed when the plane waves are replaced by spatial waves in a thin liquid film [30]). For known values of k_* and δ_*, it is not difficult to determine the vibrational frequencies $\Omega_* = \Omega''(k_*)\delta_*$ for the plane thermocapillary motion considered here, and the amplitudes of all of the variables can be determined. The theory will be approximately correct only under the condition that the amplitude of the second harmonic be much smaller than the amplitude of the principal harmonic and that the Reynolds and Peclet numbers be small in comparison to unity. In addition, the inequality (5) must be satisfied and the vibrational frequency Ω_* must be much smaller than the characteristic inverse times $\nu_j k_*^2$ and $\varkappa_j k_*^2$ of propagation of momentum and heat over a distance of order k_*^{-1}.

If the system (22) does not have real positive roots satisfying the maximum condition on γ, an ordered convective regime of the kind considered above cannot occur. This means that the nonlinear dependence (6) cannot by itself stabilize the system against increasing perturbations, and the latter will amplify, unless other nonlinearities not considered here stabilize the system. If this does not occur for finite, small-amplitude perturbations, the instability leads to vibrations with various values of k and Ω. This corresponds to a "rigid" loss of stability and interfacial turbulence.

The number of parameters affecting the characteristics of thermocapillary convection is large, so that a detailed analysis of even the simple problem considered here would be very difficult and may be the subject of a separate paper. Here we consider as an example two special cases: 1) the liquids are such that R = 0; and 2) the quantity k_* satisfies the condition $k_*^2 \ll R/S$. In both of these cases we take for simplicity c = 0; this corresponds to approximating the function (6) by a second degree polynomial.

In both cases, Eq. (22) reduces to the same form

$$\begin{aligned} &(ar - sk_*^2)(a^2r^2 + 16s^2k_*^4) - 2ab^2r^3\delta_* = 0, \\ &s(a^2r^2 + 16s^2k_*^4)^2 - 64ab^2r^3s^2k_*^2\delta_* = 0 \end{aligned} \tag{23}$$

(we assume that $k_* \neq 0$). Eliminating δ_*, we obtain the equation

$$k_*^4 - \frac{2}{3}\frac{ar}{s}k_*^2 + \frac{1}{48}\left(\frac{ar}{s}\right)^2 = 0. \tag{24}$$

This equation has two positive roots, and the following root gives the maximum γ:

$$k_* = \left[\frac{1}{3}\left(1 + \sqrt{\frac{39}{48}}\right)\frac{ar}{s}\right]^{1/2} = \left(p\frac{ar}{s}\right)^{1/2} \approx 0.796\left(\frac{ar}{s}\right)^{1/2}. \tag{25}$$

From (23) and (25) we have

$$\delta_* = \frac{(1+16p^2)^2}{64p}\left(\frac{a}{b}\right)^2 \approx 1{,}360\left(\frac{a}{b}\right)^2. \tag{26}$$

Finally, from (21) and (25), (26), we have in the first case (R = 0)

$$\Omega_* = \frac{1+16p^2}{8}\frac{ar}{S} \approx 0.928\frac{ar}{S} \tag{27}$$

and in the second case $(k_*^2 \ll R/S)$

$$\Omega_* = p\frac{1+16p^2}{8}\frac{a^2r^2}{sR} \approx 0.588\frac{a^2r^2}{sR}. \tag{28}$$

The ratio of the amplitudes of the second and first harmonics of the temperature perturbation is characterized by $A'A'^*/AA^* = 1/64p \approx 0.025$, so that this regime is in fact almost harmonic. The condition (5) leads to the inequality $a^3/b^2 \ll \beta_j s/r$, and the requirement that the Reynolds and Peclet numbers be small in comparison to unity gives $a^3/b^2 \ll \chi^2(\mu_1+\mu_2)r$, where χ stands for any of the quantities ν_j, $\varkappa_j$ (j = 1, 2). Here as a characteristic velocity we take (see (15)) the quantity $(BB^*)^{1/2} \sim a(\mu_1+\mu_2)\delta_*^{1/2}$, and as a characteristic temperature $(AA^*)^{1/2} = \delta_*^{1/2}$. The linear dimension is k_*^{-1} and we used relations (25) and (26). For finite b and r it is clear that all the inequalities can be satisfied if a is sufficiently small and the temperature gradient β_j is sufficiently large.

Finally, to simplify the calculations, it has been assumed that ω is small. This assumption implies that $\Omega_* \ll \chi k_*^2$, which reduces to $s/S \ll \chi$ in the first case mentioned above, and to $ar/R \ll \chi$ in the second, where we have used (27) and (28) for Ω_* and (25) for k_*. From the definitions of s and S in (18) and (20) it follows that in the first case these inequalities cannot be satisfied simultaneously (for all χ) and in general we must consider $s/S \sim \chi$. Therefore, the assumption that ω is small, which was essential to our calculation, in this case is not true. Nevertheless, it can be shown that our results correctly describe the nature of the dependence of k_*, δ_*, and Ω_* on the physical parameters and are correct as order of magnitude estimates. In the second case the inequality $ar/R \ll \chi$, and also the inequality $ar/R \ll s/S$, is equivalent to the condition $k_*^2 \ll R/S$, which can be satisfied for small enough r. It is clear from our simplified analysis, however, that in a more complete theory the assumption that the frequency of thermocapillary flow is small should be rejected.

Since the Peclet number was assumed to be small, the convective motion considered here cannot lead to a significant increase in heat transport across the surface. But mass transport can significantly increase as a result of the convective motion if the Schmidt number is large, as is usually the case for diffusion in liquids.

NOTATION

A, A', B, B', integration constants; a, b, c, coefficients in the expansion (6); C, specific heat capacity; k, wave number; p, numerical constant in (25); r, s and R, S, parameters in (18) and (20); t, time; v, velocity; x, z, tangential and normal coordinates; β, temperature gradient; γ, perturbation growth factor; δ, amplitude of the principal harmonic of temperature vibrations squared; ε, small parameter; $\varkappa, \lambda$, thermal diffusivity and thermal conductivity; μ, ν, dynamic and kinematic viscosity; ρ, density; σ, surface tension; τ, temperature; ψ, stream function; ω, Ω, complex and real frequencies; subscripts j = 1, 2 refer to the two liquids, an asterisk subscript refers to stationary convection, capital letters denote the coefficients in the Fourier expansions for quantities denoted by corresponding lower case letters, an asterisk superscript means the complex conjugate.

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