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On the parametric excitation of thermoelectric instability in a liquid layer open to air

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Abstract

Results are given about the parametric excitation of thermoelectric instability in a liquid semiconductor or an ionic melt layer subject to a harmonically time varying heat flux normal to its top open surface. First, it is shown that for a purely thermoelectric instability the 'integer' disturbances are the most dangerous ones for the liquid quasiequilibrium. Then the boundaries of instability and characteristics of critical disturbances are found for the cases of coupled phenomena between thermoelectric effects and surface tension gradients and thermoelectric effects and buoyancy. Both 'half-integer' and 'integer' modes were studied. Qualitative and quantitative results are provided for the experimental observation of our predictions. © 1999 Elsevier Science Ltd. All rights reserved.

1. Introduction

There are different factors which may trigger the convective instability of a liquid layer open to air. Excitation of thermal convection in molten salts and some semiconductors may be due to a thermoelectric instability. In this case the temperature gradient creates an inner electric field in the liquid and the interaction of charge density with this field may yield instability. There are studies of stationary thermoelectric convection [1–3]. Here we study the thermoelectric effects that coupled to buoyancy and surface tension gradients (Marangoni effect) may lead to an instability of a heated liquid layer giving rise to cellular motion. For illustration we take the case of a liquid semiconductor. If the thermoelectric effects are present then convection is possible when heating the top open free surface, while no instability occurs with the other two excitatory mechanisms (buoyancy and Marangoni effect).

In particular we consider the case of parametrically excited thermoelectric convection. Modulation on a driving constraint like heat transfer across the layer is expected to alter convective instability thresholds, hence, stabilize or destabilize the otherwise liquid quasiequilibrium. Controlling different instability mechanisms and their interactions allows control of the convective motions in various technological situations like processes of semiconductor production.

When a liquid layer is heated at a top open free surface thermocapillary (Marangoni) instability has been predicted for a suitable value of the thermal gradient. The problem of Marangoni instability, parametrically excited with a periodically varying thermal flux on the free surface was studied in [4,5]. In [4] the solution of the amplitude eigenvalue problem was found analytically using a Fourier method, with just a few terms. The asymptotic dependence of the critical Marangoni number on the Prandtl number was

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Nomenclature

a, b	coefficients, Eqs. (11) and (12)
B	thermoelectric parameter, $\epsilon\gamma^2\Theta^2/\rho_1\chi\nu$
c	phase velocity of the electromagnetic wave [m s^{-1}]
c_p	specific heat of the material [J kg^{-1} K^{-1}]
d	ratio deciding the balance between thermoelectricity and buoyancy
D	charge diffusion coefficient [m^2 s^{-1}]
$\boldsymbol{e}$	unit vector along the downward vertical direction
e	electric charge
$\boldsymbol{E}$	electric field [V m^{-1}]
$\boldsymbol{E}_0$	electric field distribution in quasiequilibrium
f_1	dimensionless electric field
f_2	dimensionless gradient of electric charge
f_3	dimensionless temperature gradient
g	acceleration of gravity [m s^{-2}]
h	characteristic length scale of electromagnetic effects
H	interval of integration
k	wave number
k_0	Boltzmann constant [J K^{-1}]
Ma	Marangoni number, $\alpha_1\Theta\delta_t/\eta\chi$
n_0	concentration of electric charge [m^{-3}]
p	pressure [N m^{-2}]
p'	perturbation of pressure
Pe	electrical Prandtl number, $\epsilon\omega/2\sigma$
Pr	Prandtl number, ν/χ
Q_0	amplitude of oscillatory driving heat flux
r	Debye–Hückel radius $(\epsilon k_0 T_{00}/(e^2 n_0))^{1/2}$
Ra	Rayleigh number, $g\beta\Theta\delta_t^3/\nu\chi$
t	time [s]
T	temperature [K]
T_0	temperature distribution in quasiequilibrium
T_{00}	average temperature of the liquid
$\boldsymbol{v}$	velocity vector [m s^{-1}]
w	amplitude of velocity perturbation
x, z	horizontal and vertical coordinates

Greek symbols

α	surface tension [N m^{-1}]
α_0	surface tension for $T = T_0$
α_1	temperature dependence of surface tension, $\mathrm{d}\alpha/\mathrm{d}T$ [N (m^{-1} K^{-1})]
β	thermal expansion coefficient [K^{-1}]
γ	thermoelectric constant [m kV K^{-1}]
Γ	ratio of heat and diffusion skin layer, δ_t/r
δ_t	depth of heat skin-layer, $(2\chi/\omega)^{1/2}$
Δ	Laplace operator, $\mathrm{d}^2/\mathrm{d}z^2 - k^2$
ΔS	surface area
ϵ	permittivity [F m^{-1}]
η	dynamic viscosity [kg (m^{-1} s^{-1})]
θ	amplitude of temperature perturbation
ϑ	perturbation of temperature
Θ	characteristic temperature, $Q_0\delta_t/\lambda$
κ	$1/\delta_t$
λ	heat conductivity of fluid [W (m^{-1} K^{-1})]

μ	magnetic permeability [G m^{-1}]
ν	kinematic viscosity [m^2 s^{-1}]
ρ	charge density
ρ_1	density of liquid [kg m^{-3}]
σ	electric conductivity [ohm^{-1} m^{-1}]
ϕ	auxiliary field for numerical calculation, $\phi=\Delta w$
φ	electric potential
χ	heat diffusivity [m^2 s^{-1}]
ω	frequency of heat flux oscillations [rad s^{-1}]

Subscripts

m	minimum value of parameter
x, z	x or z component
$*$	critical value

determined in two limiting cases, zero and infinite Prandtl numbers. Ref. [5] was devoted to the numerical study of the amplitude problem by means of a finite-difference method. The competition between 'half-integer' and 'integer' modes was investigated. It was shown that in the region of a high Prandtl number ($Pr > Pr_* = 1.2$) the 'half-integer' mode is responsible for instability, while in the region of a low Prandtl number ($Pr < Pr_*$) the 'integer' mode is the most dangerous. The role of finite thickness of the layer, as well as that of Newtonian heat transfer on the free surface and the interaction of the Marangoni effect (thermocapillarity) and buoyancy (thermogravitation) leading to instability were also studied in [5]. Finally, in [6] the influence of the deformation of the free surface on the Marangoni instability was discussed where there is parametric heat driving.

In the perspective of the earlier mentioned studies here we discuss the dynamic excitation of thermoelectric convection relative to the also possible excitation due to thermocapillary and thermogravitational effects when these two are parametrically driven. The description of the problem is given in Section 2. The method of the solution is discussed in Section 3. Convective thresholds for different modes of instability are presented in Section 4, together with estimates amenable to experimental test.

2. Description of the problem

We consider a semi-infinite liquid layer, $0 < z < \infty$, with an open top, undeformable level surface. The z-axis is directed vertically downwards. The layer is heated in such a way that the uniform heat flux periodically varies with time along the surface and is directed normally to the initially flat free surface that we consider located at $z = 0$:

$$\lambda\left(\frac{\partial T}{\partial z}\right)_0 = Q_0 \cos \omega t \tag{1}$$

where λ is the heat conductivity of fluid, Q_0 and ω are, respectively, the amplitude and the frequency of the oscillatory driving heat flux.

For illustration we consider a semiconductor liquid or an ionic melt such that the temperature gradient, ∇T, creates an inner electric field, $\boldsymbol{E} = \gamma\nabla T$, due to thermodiffusion of free electric charges, with γ denoting the thermoelectric coefficient. However, we assume that the electric conductivity of the liquid is low enough in order to have a negligible magnetic field, which is expected to be induced by the electric current. For simplicity we also neglect the Joule heat in the energy equation and both the nonuniform polarisation and the nonuniform conductivity of the liquid. Then the fields describing the possible convection in the liquid are velocity $\boldsymbol{v}$, pressure p, temperature T, charge density ρ, electric $\boldsymbol{E}$ and electrical potential φ. Under the earlier given approximations these fields obey the following equations:

$$\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v}\nabla)\boldsymbol{v} = -\frac{1}{\rho_1}\nabla p + \nu\nabla^2\boldsymbol{v} - g\beta T\boldsymbol{e} + \rho\boldsymbol{E}, \quad \operatorname{div}\boldsymbol{v} = 0$$

$$\frac{\partial T}{\partial t} + (\boldsymbol{v}\nabla)T = \chi\nabla^2 T$$

$$\frac{\partial \rho}{\partial t} + (\boldsymbol{v}\nabla)\rho = -\operatorname{div}(\sigma\boldsymbol{E} - D\nabla\rho - \sigma\gamma\nabla T)$$

$$\epsilon \operatorname{div}\boldsymbol{E} = \rho, \quad \boldsymbol{E} = -\nabla\varphi$$

$$\boldsymbol{e} = (0,0,1) \tag{2}$$

where ρ_1, ν, χ, β, σ, ϵ are, respectively, the liquid density, the kinematic viscosity, the heat diffusivity, the thermal expansion coefficient, the electric conductivity and the permittivity. D is the charge diffusion coefficient. Note that due to the approximations invoked we remain in the EHD realm, thus neglecting all magnetic field effects. Indeed in our study we consider that

$$\sigma \ll \frac{1}{h}\left(\frac{\epsilon}{\mu}\right)^{1/2}, \quad \frac{\omega}{2\pi} \ll \frac{c}{h} \tag{3}$$

where μ is the magnetic permeability, c is the phase velocity of the electromagnetic wave in the liquid and h is a characteristic length scale of electromagnetic effects. Typical values of the parameters we consider are

$$\epsilon \sim 88.5 \cdot 10^{-12}\ \mathrm{F\,m^{-1}}, \quad \mu \sim \mu_0 = 4\pi \cdot 10^{-7}\ \mathrm{G\,m^{-1}},$$

$$h \sim 10^{-1}\ \mathrm{m}, \quad c \sim 10^{8}\ \mathrm{m\,s^{-1}},$$

$$\sigma \ll 0.1\ \mathrm{om^{-1}\,m^{-1}}, \quad \omega \ll 10^{9}\ \mathrm{rad\,s^{-1}}. \tag{4}$$

Liquids satisfying these conditions are ionic melts such as BeF_2, $HgCl_2$, AlI_3 ($\sigma \backsim 10^{-4}$–10^{-3} $\mathrm{om^{-1}\ m^{-1}}$) [7] or liquid semiconductors like Se ($\sigma \backsim 10^{-4}$ $\mathrm{om^{-1}\ m^{-1}}$) [8].

As a base state we consider that the liquid layer is in mechanical motionless quasiequilibrium. The temperature field $T_0(z, t)$ satisfies the one-dimensional heat diffusion equation:

$$\frac{\partial T_0}{\partial t} = \chi \frac{\partial^2 T_0}{\partial z^2} \tag{5}$$

At the open top surface we have the thermal boundary condition (1). Far away down in the liquid layer, the temperature is chosen as the reference temperature:

$$(T_0)_{z\to\infty} = 0. \tag{6}$$

Accordingly, the temperature and temperature gradient become:

$$T_0 = -\frac{Q_0}{2\lambda\kappa}[\cos(\omega t - \kappa z) + \sin(\omega t - \kappa z)]\exp(-\kappa z),$$

$$\frac{\partial T_0}{\partial z} = \frac{Q_0}{\lambda}\cos(\omega t - \kappa z)\exp(-\kappa z), \quad \kappa = \sqrt{\frac{\omega}{2\chi}}. \tag{7}$$

The solution of the heat diffusion equation is a heat wave propagating from the top surface downwards, damped inside the liquid and with a temperature 'skin-layer' of characteristic length scale, $\delta_t = 1/\kappa$.

To formulate the problem in non-dimensional form we introduce the following scales: $\delta_t = 1/\kappa$, for length; $\delta_t^2/\chi = 2/\omega$, for time; χ/δ_t, for velocity; $\rho_1\chi\nu/\delta_t^2$, for pressure; $\Theta = Q_0\delta_t/\lambda$, for temperature; $\epsilon\gamma\Theta/\delta_t^2$, for electrical charge; $\gamma\Theta$, for electric potential; and $\gamma\Theta/\delta_t$, for electric field. Then the system (2) becomes

$$\frac{1}{Pr}\left(\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v}\nabla)\boldsymbol{v}\right) = -\nabla p + \nabla^2 \boldsymbol{v} - Ra\, T\boldsymbol{e} + B\rho\boldsymbol{E}$$

$$\frac{\partial T}{\partial t} + (\boldsymbol{v}\nabla)T = \nabla^2 T$$

$$Pe\left(\frac{\partial \rho}{\partial t} + (\boldsymbol{v}\nabla)\rho\right) = -\rho + \frac{1}{\Gamma^2}\nabla^2\rho + \nabla^2 T \tag{8}$$

$$\operatorname{div}\boldsymbol{v} = 0, \quad \operatorname{div}\boldsymbol{E} = \rho, \quad \boldsymbol{E} = -\nabla\varphi$$

$$B = \frac{\epsilon\gamma^2\Theta^2}{\rho_1\chi\nu}, \quad Ra = \frac{g\beta\Theta\delta_t^3}{\nu\chi}, \quad \Gamma^2 = \left(\frac{\delta_t}{r}\right)^2$$

$$P = \frac{\nu}{\chi}, \quad Pe = \frac{\epsilon\omega}{2\sigma} = \frac{\epsilon\chi}{\sigma\delta_t^2} \tag{9}$$

where $r = \sqrt{\epsilon k_0 T_{00}/(e^2 n_0)}$ is the Debye–Hückel radius, k_0 is Boltzmann's constant, T_{00} is a mean reference temperature in the liquid layer, e is the electric charge, n_0 is the concentration of electric charges, Pr is the Prandtl number, Pe is the electric Prandtl number, B is the thermoelectric parameter, and Ra is the Rayleigh number.

As already said we assume that the electroconductivity is of negligible influence on the evolution of the liquid layer and that any excess charge is created only by thermodiffusion and diffusion, hence $Pe \approx 0$. For illustration note that for liquid Se, $\gamma = 10^3$ mkV $\mathrm{K^{-1}}$, $\sigma \backsim 10^{-4}$ $\mathrm{om^{-1}\ m^{-1}}$ [8]; for frequencies $\omega < 10^2$ rad $\mathrm{s^{-1}}$ yields $Pe \approx 0$.

In quasiequilibrium, i.e. when liquid motion is negligible, the charge distribution $\rho_0(z, t)$ and the electric field $\boldsymbol{E} = (0, 0, E_0(z, t))$ are given by

$$\frac{1}{\Gamma^2}\cdot\frac{\partial^2\rho_0}{\partial z^2} - \rho_0 = -\frac{\partial^2 T_0}{\partial z^2}$$

$$\frac{\partial E_0}{\partial z} = \rho_0 \tag{10}$$

with T_0 given by (7). Then the charge density and the electric field are

$$E_0 = \exp(-\Gamma z)\left(\frac{b-a}{2}\ (2t) - \frac{a+b}{2}\sin(2t)\right)$$

$$+ \exp(-z)\left(\frac{a-b}{2}\cos(2t - z) - \frac{a+b}{2}\sin\right. \tag{11}$$

$$\left.(2t - z)\right).$$

$$\rho_0 = \Gamma \cdot \exp(-\Gamma z)\left(\frac{a-b}{2}\cos(2t) + \frac{a+b}{2}\sin(2t)\right) + \exp(-z)(a\sin(2t-z) + b\cos(2t-z)). \tag{12}$$

$$a = \frac{1+2/\Gamma^2}{1+4/\Gamma^4}, \quad b = \frac{-1+2/\Gamma^2}{1+4/\Gamma^4}. \tag{13}$$

These distributions contain two different terms coming, respectively, from the diffusion boundary layer and the heat boundary layer. Here we restrict consideration to the case when the former is much shallower than the latter, i.e., $\Gamma \gg 1$, and, subsequently, we disregard the diffusive part of all distributions. Accordingly, we do not take into account the influence of the diffusion boundary layer on the dynamics of the liquid. The order of the differential equation which describes the charge density disturbances is lowered relative to the general case. Furthermore, the evolution of the charge is determined only by the thermoelectric effects. Eventually, the problem reduces to the investigation of the dielectric-like liquid. If $(1/\Gamma^2) \ll 1$ then $a=1$, $b=-1$, $\exp(-\Gamma z)$ tends to zero and we have

$$E_0 = \exp(-z)\cos(2t-z) \tag{14}$$

$$\rho_0 = \exp(-z)(\sin(2t-z) - \cos(2t-z)). \tag{15}$$

We shall investigate the role of the Prandtl number on the onset of instability. Our assumption about the thickness ratio of diffusive and thermal boundary layers ($\Gamma \gg 1$) is still acceptable at arbitrary Prandtl numbers. Indeed, this dimensionless parameter characterises the ratio of viscous and thermal boundary layers, $Pr = \nu/\chi = (2\nu/\omega)\cdot(\omega/2\chi) = (\delta_\nu/\delta_t)^2$ and does not influence Γ.

Now we consider the stability of the motionless mechanical quasiequilibrium (4). Let $\boldsymbol{v}$, ϑ, p', ρ, $\boldsymbol{E}$, φ, denote small disturbances on the temperature, velocity, pressure, electric charge and electric field, respectively. From Eqs. (2) follows that these disturbances obey the linearized Navier–Stokes, continuity, heat transfer, charge density and electrostatic equations near the quasiequilibrium solution:

$$\frac{1}{Pr}\frac{\partial \boldsymbol{v}}{\partial t} = -\nabla p' + \nabla^2 \boldsymbol{v} - Ra\,\vartheta\cdot\boldsymbol{e} + B(\rho_0 \boldsymbol{E} + \rho \boldsymbol{E}_0)$$

$$\frac{\partial \vartheta}{\partial t} + (\boldsymbol{v}\nabla)T_0 = \nabla^2\vartheta$$

$$\rho = \nabla^2\vartheta$$

$$\operatorname{div}\boldsymbol{v} = 0, \quad \operatorname{div}\boldsymbol{E} = \rho, \quad \boldsymbol{E} = -\nabla\varphi \tag{16}$$

together with the appropriate boundary conditions. As the normal heat flux on the top flat free surface is prescribed by the condition (1), we assume that disturbances on the prescribed temperature gradient at the open surface vanish

$$\left(\frac{\partial \vartheta}{\partial z}\right)_{z=0} = 0. \tag{17}$$

As there is no net charge

$$\int \rho\,\mathrm{d}V = 0 \tag{18}$$

the Gauss–Ostrogradsky theorem yields

$$\int \rho\,\mathrm{d}V = \int \operatorname{div}\boldsymbol{E}\,\mathrm{d}V = \int \boldsymbol{E}\,\mathrm{d}\boldsymbol{S} = 0. \tag{19}$$

We take the distance between the lateral areas to be equal to the spatial periodicity of the disturbances, hence,

$$\int \boldsymbol{E}\mathrm{d}\boldsymbol{S} = (E_z\Delta S_1)_{z=0} + (E_z\Delta S_2)_{z=\infty} + (E_x\Delta S)_{\text{side}} = 0 \tag{20}$$

where E_z and E_x are the components of electric field disturbances on the surface areas. ΔS are the surfaces of these areas. As disturbances tend to zero at large distances from the upper open boundary the second term in (20) vanishes. Periodicity of disturbances gives zero for the third terms in (20). Finally, we have

$$E_z = 0, \quad \left(\frac{\partial \varphi}{\partial z}\right)_{z=0} = 0. \tag{21}$$

The normal component of velocity is zero at open surface:

$$(v_z)_{z=0} = 0. \tag{22}$$

Let us now examine the possibility of surface tension gradient-driven or thermocapillary (Marangoni) instability. We take the surface tension, α, linearly varying with temperature:

$$\alpha = \alpha_0 - \alpha_1 T \tag{23}$$

with α_1 positive. The tangential stress balance of viscous and thermocapillary forces is

$$\frac{\partial^2 v_z}{\partial z^2} = -Ma\frac{\partial^2 \vartheta}{\partial x^2} \tag{24}$$

where $Ma = \alpha_1\Theta\delta_t/\eta\chi = 2\alpha_1 Q_0/\eta\lambda\omega$ is the Marangoni number and η is the dynamic viscosity.

Our problem is isotropic in the plane of the liquid surface, hence all disturbances depend on a single coordinate, here x. As far away from the free surface all disturbances decay to zero we have

$$(\boldsymbol{v},\vartheta,\rho,\boldsymbol{E},\varphi)_{z\to\infty}\longrightarrow 0. \tag{25}$$

Eliminating the horizontal velocity component, v_x, the pressure disturbances p', and the charge density, ρ, we can formulate the problem in terms of the normal velocity component, v_z, and temperature disturbances, θ. Indeed, assuming that

$$\begin{pmatrix} v_z \\ \vartheta \\ \rho \\ \boldsymbol{E} \\ \varphi \end{pmatrix} = \begin{pmatrix} w(z,t) \\ \theta(z,t) \\ \rho(z,t) \\ \boldsymbol{E}(z,t) \\ \varphi(z,t) \end{pmatrix} \exp\,[ikx] \tag{26}$$

where k is the wave number, we obtain the boundary value problem for the amplitudes w, θ, ρ, φ.

$$\frac{1}{Pr}\frac{\partial \Delta w}{\partial t} = \Delta^2 w + Ra\,k^2\theta - Bk^2(\rho f_1 + \varphi f_2),$$

$$\Delta = \frac{\mathrm{d}^2}{\mathrm{d}z^2} - k^2$$

$$\frac{\partial \theta}{\partial t} + f_3 w = \Delta\theta$$

$$\Delta\varphi = -\rho, \quad \rho = \Delta\theta. \tag{27}$$

$$z = 0:\quad \theta' = 0, \quad \varphi' = 0, \quad w = 0, \quad w'' = k^2\,Ma\,\theta$$

$$z \longrightarrow \infty:\quad \theta = 0, \quad \varphi = 0, \quad w = 0, \quad w' = 0. \tag{28}$$

Here prime denotes differentiation with respect to the vertical coordinate z; f_1 is the electric field E_0 (14); $f_2 = \partial\rho_0/\partial z = -2\,\sin(2t-z)\,\exp(-z)$ is the gradient of electric charge; $f_3 = \cos(2t-z)\,\exp(-z)$ is the dimensionless temperature gradient in the liquid. All are time-dependent functions that define the quasiequilibrium. Then for w and θ we have

$$\frac{1}{Pr}\frac{\partial \Delta w}{\partial t} = \Delta^2 w + Ra\,k^2\theta - Bk^2(\Delta\theta f_1 - \theta f_2)$$

$$\frac{\partial \theta}{\partial t} + f_3 w = \Delta\theta \tag{29}$$

$$z = 0:\quad \theta' = 0, \quad w = 0, \quad w'' = k^2\,Ma\,\theta$$

$$z \longrightarrow \infty:\quad \theta = 0, \quad w = 0, \quad w' = 0. \tag{30}$$

The system of equations, boundary conditions (29) and (30) and the condition of periodicity in time for all the variables define the eigenvalue problem for B or Ma as a function of the remaining parameters like the Marangoni number, the Prandtl number, the Rayleigh number and the wavenumber. Two periodic solutions are of interest: one with a period twice the period of the heat flux on the boundary ('half-integer' solutions) and the other with the same period as the heat flux ('integer' solutions). The minimum on the neutral stability curve of the function $B(k)$, for fixed values of all other parameters, determines the critical value, B_{m}, for instability due to the parametric excitation in the liquid layer.

Note that the problem (29) and (30) besides invariance under the standard symmetry transformations: (i) $t \to t+2\pi$, $w \to w$, $\theta \to \theta$, $\rho \to \rho$ for the 'half-integer' mode, and (ii) $t \to t+\pi$, $w \to w$, $\theta \to \theta$, $\rho \to \rho$ for the 'integer' mode, it also has the symmetry:

$$t \longrightarrow t + \pi/2, \quad w \longrightarrow -w, \quad \theta \longrightarrow \theta, \quad \rho \longrightarrow \rho. \tag{31}$$

This solution belongs to a subset of the 'integer' solutions.

3. Solution of the problem

Periodic solutions of the eigenvalue problem have been found with the help of a finite-difference method, on a finite interval of z: $0 < z < H$. The layer depth H was taken larger than the temperature wave skin-layer thickness δ_t, hence $H/\delta_t \gg 1$. It appears that the critical parameters (for the thermoelectric number, for instance) remain practically constant when the ratio H/δ_t increases from 10 to 15. In the considered region, a uniform grid with a constant step $h_z = 0.1$ was used (test calculations were carried out with the step $h_z = 0.05$). The auxiliary variable $\phi = \Delta w$ has been used. The problem was solved in terms of the variables w, ϕ, θ. The boundary condition at $z = H$ corresponds to a rigid isothermal plane. For ϕ we have a condition analogous to the Toms condition, and on the free surface $\phi = 0$.

An implicit finite-difference scheme was used. In constructing the finite difference analog of the convective equations, we approximated the derivatives with respect to the spatial coordinate and time using central and forward differences, respectively. The corresponding algebraic system of equations has been solved by the sweep method. The time step remained constant and was selected to ensure stability and the prescribed accuracy in the determination of the critical thermoelectric number or the Marangoni number.

The following procedure to obtain the critical value of the thermoelectric parameter was used. For given parameter values of Pr, k and Ma, an initial local temperature disturbance or a velocity field disturbance were introduced in the inner point on the coordinate grid (usually at $z = 1$). For arbitrary B the solution

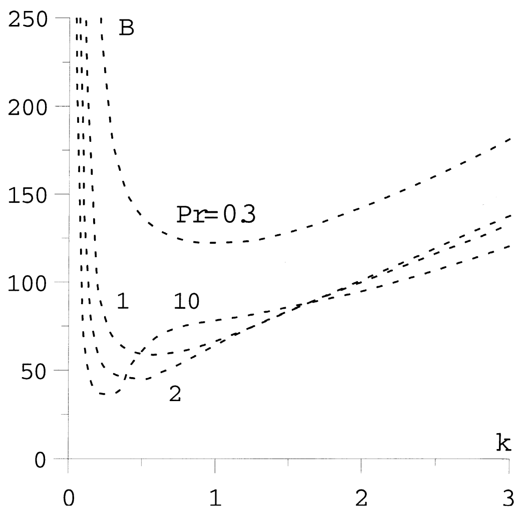

Fig. 1. Neutral stability curves for pure thermoelectric convective instability, $B(k)$; $Ma=0$; $Ra=0$; $Pr=0.3$, 1, 2, 10.

either grows or decays with time. This can be determined from the value of the multiplier (the logarithm of the ratio velocity disturbances at times differing by a period). The value B corresponding to the neutral solution was obtained for zero value of the multiplier. Thus, we have constructed the neutral curve, $B(k)$, of the critical disturbances in terms of the wavenumber k. For fixed Pr and Ma, minimization of the function $B(k)$ gives the threshold value, B_{m}, for the dynamic excitation of thermoelectric convection.

The behaviour of disturbances of different types was studied. The period of the first ('half-integer' mode) exceeded the period of modulation of the external heat flux by a factor of two. The period of the other ('integer' mode) was equal to the period of the external drive. At some parameter values, the solutions with symmetry (31) were observed. Also, for certain values of the parameters (in the neighbourhood of the neutral curves) we carried a Fourier analysis of the velocity and temperature disturbances. We, indeed, identified the disturbances as either a 'half-integer' or 'integer' type.

4. Quantitative and qualitative results amenable to experimental test

Some numerical results are presented in Figs. 1–6. The solid curves correspond to the stability boundary for 'half-integer' disturbances, while the dashed lines correspond to the boundary for 'integer' ones. First, we consider the pure thermoelectric case ($Ma=0$; $Ra=0$). For a few Prandtl numbers the neutral curves

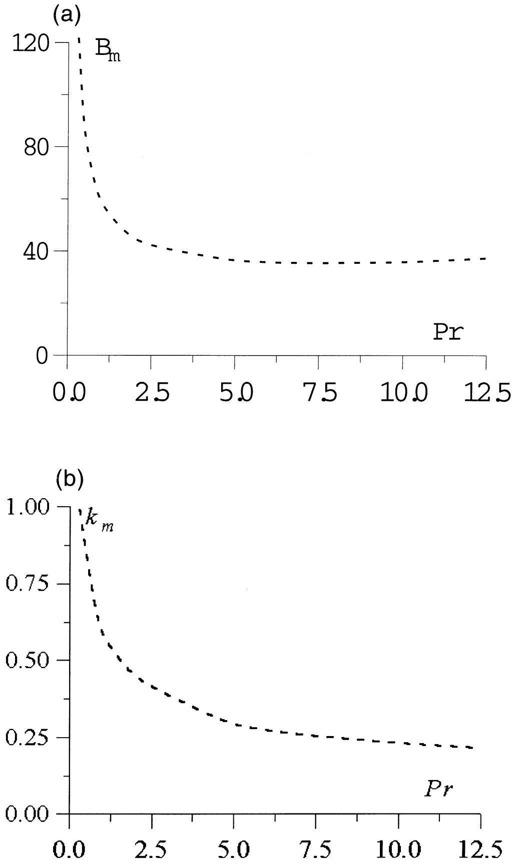

Fig. 2. Critical parameters of pure thermoelectric instability vs Prandtl number: (a) threshold of convection $B_{\mathrm{m}}(k)$; (b) wavenumber k_{m}.

are presented in Fig. 1. For $Pr=1$ the critical value of the thermoelectric number $B_{\mathrm{m}}=58.81$ corresponds to a critical wavenumber $k_{\mathrm{m}}=0.58$. 'Integer' periodic disturbances grow in the domains under the curves. This is the convective parametric mode of the thermoelectric instability. Contrary to the case of parametric excitation of buoyancy-driven, thermogravitational convection [9–11] or the problem of dynamic thermocapillary Marangoni instability [4–6], only 'integer' disturbances are possible for the purely thermoelectric case. Note that as the thermoelectric parameter is proportional to the square of the temperature gradient, then varying its direction does not change the sign of the effect as the sign of the Coulomb force in the Navier–Stokes equations remains unaltered.

The dependence of the critical thermoelectric number, B_{m}, and critical wavenumber, k_{m}, on the Prandtl number in the pure thermodynamic case ($Ma=0$; $Ra=0$) are presented in Fig. 2. Both the critical thermoelectric number and the wavenumber decreases as

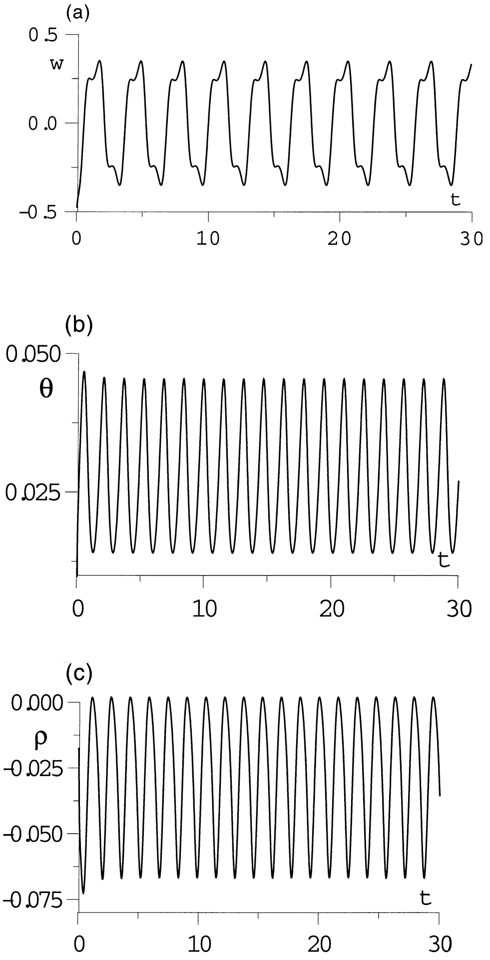

Fig. 3. Time evolution of critical disturbances in pure thermoelectric convective instability: (a) w, velocity; (b) θ, temperature; and (c) ρ, charge density: $Ma=0$; $Ra=0$; $Pr=10$; $k=0.75$.

the Prandtl number increases. When $Pr \gg 1$ the figure provides values that agree well with the independently obtained asymptotic values $B_{\mathrm{m}} \sim$ const; $k_{\mathrm{m}} \sim$ const. Indeed, in the limit of a high Prandtl number the asymptotic behaviour can be found using a scaling approach. In this limit the inertial term in the momentum equations can be neglected, therefore, $B=$const and $k=$const, as the critical thermoelectric number and wavenumber do not depend on Pr. This behaviour of the critical values is analogous to the result found for the pure thermocapillary (Marangoni) and the pure thermogravitational, buoyancy-driven cases [4,11]. The time-dependence of the critical disturbances which corresponds to the symmetry transformation (31) is shown in Fig. 3. The time period of the charge density and temperature disturbances is $\pi/2$, while for the velocity it is π.

Competition between the ‘half-integer’ and ‘integer’ instability modes is possible if we consider the interaction between the two mechanisms of instability; for instance, thermoelectricity and thermocapillarity and thermoelectricity and buoyancy. The dependence of the Marangoni number on the thermoelectric parameter B (minimizing with respect to the wavenumber k) is presented in Fig. 4 for Prandtl number $Pr=1$. The stability domain is below the curves. Curves I and II are the instability boundaries for thermoelectric and thermocapillary (Marangoni) convection, respectively. For Marangoni instability ($B=0$) we have: $Ma=110$ for the ‘half-integer’ type, and $Ma=295$ for the integer’ one. Increasing the thermoelectric parameter only slightly affects the ‘half-integer’ mode of instability while it strongly influences the ‘integer’ one. This behaviour is indeed expected as the thermoelectricity excites only the ‘integer’ mode.

The neutral curves $B(k)$ for $Ma=131$ are presented in Fig. 5. The thermoelectric instability domain is above the ‘I’ curve, while the thermocapillary (Marangoni) instability domain is below the ‘II’ curve. There is a very narrow stability region, $51.96 < B < 53.58$.

For a few Prandtl numbers the dependence of the critical Marangoni number on the thermoelectric parameter is presented in Fig. 6. The growth of the Prandtl number decreases the stability region. At high enough Prandtl numbers the two mechanisms of instability practically do not influence one another.

The competition of thermogravitation (buoyancy) and thermoelectricity is shown in Fig. 7 ($Ma=0$) for a few values of the Prandtl number. The stability region is inside the curves. The pure buoyancy-driven parametric instability for $B=0$ is of ‘half-integer’ type [11] and the thermoelectricity does not change its nature. It

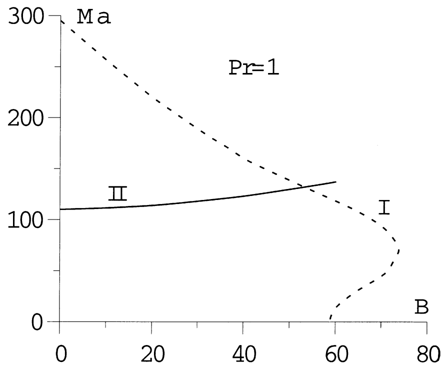

Fig. 4. Critical Marangoni number, Ma vs thermoelectric number, B, for Prandtl number $Pr=1$ when there is no buoyancy, $Ra=0$. The solid line is the stability boundary for ‘half-integer’ modes. The broken line is the stability boundary for ‘integer’ modes.

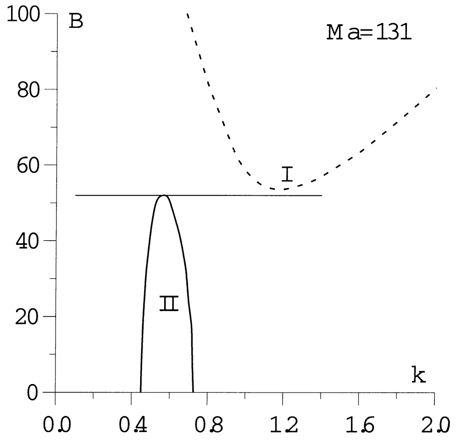

Fig. 5. Instability domains in the plane $B(k)$: $Pr=1$; $Ma=131$; $Ra=0$: I, pure thermoelectric instability; and II, pure thermocapillary Marangoni instability. Note that curve II is such that if $B=0$ there is instability for $Ma>110$. Then for $Ma=131$ we have a band of unstable modes due to thermocapillarity (Marangoni effect). Increasing B diminishes this band.

merely shifts the instability threshold (solid lines). The drastic bending on the line for the 'integer' thermoelectric model, for the relatively high Prandtl number $Pr=7$, corresponds to a transition from one to other mode of instability that correspond to different critical wavenumbers.

Finally, let us estimate some numerical values amenable to experimental test. We can take a characteristics temperature difference of a few hundred degrees like $\Theta\sim(3\text{–}4)\cdot10^2$ K, a frequency $\omega \leqslant 100$ rad s^{-1}, and for the properties of the liquid [1,7,8]:

$$\rho\sim2\cdot10^3\ \mathrm{kg\ m^3},\quad c_\mathrm{p}\sim2\cdot10^3\ \mathrm{J\ (kg^{-1}\ K^{-1})},\quad \beta\sim10^{-4}\ \mathrm{K^{-1}}$$

$$\eta\sim10^{-4}\text{–}10^{-3}\ \mathrm{kg\ (m^{-1}\ s^{-1})},\quad \epsilon\sim88.5\cdot10^{-12}\ \mathrm{F\ m^{-1}}$$

$$\sigma\sim10^{-4}\ \mathrm{om^{-1}\ m^{-1}},\quad \gamma\sim10^3\ \mathrm{mkV\ K^{-1}},\quad \lambda\sim10^{-2}\ \mathrm{W\ (m^{-1}\ K^{-1})} \tag{32}$$

where c_p is the specific heat. Then the depth of the temperature skin-layer is

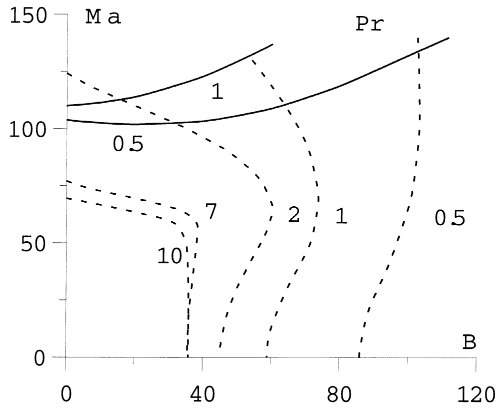

Fig. 6. Critical Marangoni number, Ma vs thermoelectric number, B, for a few Prandtl numbers, $Pr=0.5$, 1, 2, 7, 10, when there is no buoyancy, $Ra=0$. Solid and broken lines are as in Fig. 4.

$$\delta_t=\left(\frac{2\chi}{\omega}\right)^{1/2}=\left(\frac{2\lambda}{\omega c_\mathrm{p}\rho_1}\right)^{1/2}=\left(\frac{2\cdot10^{-2}}{2\cdot10^3\cdot2\cdot10^3}\right)^{1/2}\omega^{-1/2}\sim10^{-4}\omega^{-1/2}\ (\mathrm{m}) \tag{33}$$

i.e. 10^{-5} m for $\omega=100$ rad s^{-1}, and 10^{-4} m for $\omega=1$ rad s^{-1}. These estimates and the conductivity value fit well with the EHD approximation (3). Moreover, the electric Prandtl number is

$$Pe=\frac{\epsilon\omega}{2\sigma}=\frac{88.5\cdot10^{-12}}{2\cdot10^{-4}}\omega\sim5\cdot10^{-7}\omega,\quad Pe\approx0 \tag{34}$$

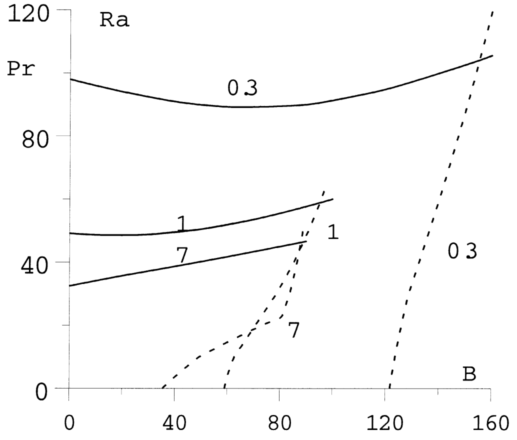

Fig. 7. Critical Rayleigh number, Ra vs thermoelectric number, B, for a few Prandtl numbers, $Pr=0.3$ 1, 7, when there is no thermocapillarity, $Ma=0$. Solid and broken lines are as in Fig. 4.

which also complies with the assumptions of our theory. Then

$$B = \frac{\epsilon\gamma^2\Theta^2}{\rho_1\chi\nu} = \frac{88.5\cdot 10^{-12}\cdot 10^{-6}}{10^{-4}\cdot 10^{-8}}\Theta^2 \sim 10^{-4}\cdot\Theta^2 \quad (35)$$

that for $\Theta \sim (3\text{–}4)\cdot 10^2$ gives $B \sim 10\text{–}16$. A slight increase in the values of Θ and ϵ bring us to the unstable region with the right order of magnitude in B.

If we disregard the Marangoni effect ($Ma=0$) and restrict consideration to the competition between thermoelectricity and buoyancy, the ratio deciding the balance between them is

$$d = \frac{B}{Ra} = \frac{\epsilon\gamma^2\Theta}{\rho_1 g\beta\delta_t^3} = \frac{88.5\cdot 10^{-12}\cdot 10^{-6}\cdot 10^{12}}{2\cdot 10^3\cdot 10^{-4}}\cdot\frac{\omega^{3/2}\Theta}{g} \sim 10^{-4}\frac{\omega^{3/2}\Theta}{g}. \quad (36)$$

Actually, $d \sim 3$ for $\omega = 100$ rad s^{-1}, and $\Theta = 300$ K on Earth ($g = 9.8$ m s^{-2}), while $d \gg 1$ for reduced gravity conditions ($g_{\text{eff}} = 10^{-4}\cdot g$).

5. Conclusions

The effect of a variable periodic heat flux on the top open undeformable surface of a layer of liquid semiconductor or ionic melt leading to instability of the initial liquid quasiequilibrium has been investigated. Domains of parametric excitation due to the resonance between the imposed heat wave and spontaneous perturbations inside the liquid were delineated. The thresholds of parametric convective instability were found for various cases of instability: purely thermoelectric instability; coupled thermoelectric and thermocapillary instability; coupled thermoelectric and buoyancy-driven instability.

For purely thermoelectric convection the ‘integer’ perturbations are shown to be the most dangerous as the Coulomb force does not depend on the direction of the temperature gradient and, consequently, does not vary its direction over a period of the external drive. In the limit of high Prandtl numbers the critical thermoelectric number and critical wavenumber become constants.

When both Marangoni and Rayleigh numbers do not vanish then results depend on the direction of the imposed temperature gradient. ‘Integer’ and ‘half-integer’ increasing disturbances are generated when there is parametric excitation of convection. The interaction between the two mechanisms of instability leads to competition between the ‘integer’ and ‘half-integer’ modes. Numerical estimates amenable to experimental test are provided for typical cases of liquid semiconductors and ionic melts.

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