

Thermoelectric convection in a liquid layer in the presence of a longitudinal temperature gradient

E. D. Eidel'man

St. Petersburg Chemical-Pharmaceutical Institute, 197376 St. Petersburg, Russia

(Submitted 19 December 1995; resubmitted 19 March 1996)

Zh. Éksp. Teor. Fiz. **110**, 891–898 (September 1996)

In the process of melting of a semiconductor (semimetal) by, for example, laser radiation, heating also occurs along the surface of the melt. We consider a model in which the heat flux is strictly parallel to the layer and take into account the thermoelectric effect. It is shown that the usual longitudinal motion under such conditions can be converted into cellular motion. The possibility of observing this effect is discussed. © 1996 American Institute of Physics. [S1063-7761(96)00909-2]

1. INTRODUCTION

In the process of obtaining semiconductor films,^{1,2} heat fluxes with a component along the layer are used. The problem in which the temperature gradient is parallel to the heated layer has been solved assuming the absence of a thermoelectromotive force.^{3–5} Flow along the layer occurs in this case with a velocity proportional to the magnitude of the temperature gradient,³ i.e., there is no threshold. This type of flow is stable (see Ref. 6 and the literature cited there), since the Rayleigh–Bénard and the Marangoni convection mechanisms require a preferred direction of heating. For example, in the Rayleigh–Bénard mechanism, cellular motion occurs only in the case of heating from below, i.e., when the temperature gradient is opposite the force of gravity. In the Marangoni mechanism, cellular motion occurs only for heating from the bottom of the liquid and does not occur when the heating is from the free surface.

The theory of the mechanism of another mode of convection in liquid semiconductors—thermoelectric convection—was developed in Refs. 7 and 8. It was shown that for a sufficiently thin layer of weakly-conducting liquid, cellular motion can occur for a sufficiently large heat flux perpendicular to the layer, independently of the direction of the heat flux with respect to the force of gravity. Physically, this type of cellular motion occurs as follows. A charge fluctuation produced by the temperature perturbation is acted upon by the thermoelectric field created by the heating. The resulting electric force results in cellular motion.

In the present paper we consider the effect of the thermoelectric force on motion in a layer produced by a heat flux parallel to the layer. It is shown that the motion produced by a heat flux directed along the layer increases without limit at the same flux responsible for cellular motion when the heat flux is perpendicular to the layer. Therefore, motion perpendicular to the layer is induced. Because the electric force mechanism of cellular motion is not associated with a specific direction in space, and the conditions of heating with a longitudinal heat flux are sufficient, it follows that all of the conditions for cellular motion are satisfied.^{7,8}

2. STATEMENT OF THE PROBLEM

We consider a liquid layer (the x axis is along the layer and the z axis is perpendicular to it) along which there is an

external constant heat flux, i.e., the temperature gradient $|\nabla T_0| = A_x = A$ due to external heating lies exactly parallel to the layer. No equilibrium state exists in the presence of a longitudinal temperature gradient, and motion along the layer occurs for arbitrarily small heating A . We consider the middle region of the layer, i.e., the region not close to the walls of the container, which are maintained at constant temperature. At the “cold” wall at $x=0$ we assume that $T=T_0=T_c=0$. In treating the middle of the layer we can assume that the temperature at the bottom and at the surface varies linearly:

$$T = Ax \quad \text{at } z=0, \quad z=h. \quad (2.1)$$

It will be shown below that A is constant throughout the liquid (for $0 \leq z \leq h$). In the middle of the layer the velocity of the liquid is such that $v = v_x \gg v_z$. We assume that $v_z = 0$, our most restrictive assumption. It will obviously break down when the temperature of the “hot” wall is sufficiently high.

The continuity equation for an incompressible liquid implies that the flow velocity depends only on z . Because the flow occurs with zero threshold (no equilibrium state), perturbation theory cannot be used, and the nonlinear equations of hydrodynamics must be used to describe the motion. For thin layers, in which the dimensions along the layer $\lambda \gg h$, the x and z components of the Navier–Stokes equations (equations of motion) can be written in the form

$$v \frac{\partial^2 v}{\partial z^2} - \frac{1}{\rho} \frac{\partial p}{\partial x} = - \frac{en\gamma}{\rho} \frac{\partial T}{\partial x}, \quad (2.2)$$

$$- \frac{1}{\rho} \frac{\partial p}{\partial z} + g\beta T = - \frac{en\gamma}{\rho} \frac{\partial T}{\partial z}. \quad (2.3)$$

These equations include the effect of gravity (g is the acceleration of gravity) and the electric force on the charge en (n is the number density and e is the charge of a carrier) produced by the thermoelectric field $\mathbf{E} = \gamma \nabla T$.

The equations of motion must be supplemented with Fourier's law of thermal conduction

$$v \frac{\partial T}{\partial x} = \kappa \left(\frac{\partial^2 T}{\partial z^2} + \frac{\partial^2 T}{\partial x^2} \right) \quad (2.4)$$

and Poisson's equation of electrostatics. Since the electric field is thermoelectric in origin, this equation has the form

$$en = \gamma \varepsilon \left(\frac{\partial^2 T}{\partial z^2} + \frac{\partial^2 T}{\partial x^2} \right). \quad (2.5)$$

We have also introduced the density of the liquid ρ , its pressure p , the kinematic viscosity ν , the thermal conductivity κ , the dielectric constant ε , the thermoelectromotive force γ , and the coefficient of volume expansion β .

Using the equations of heat conduction and electrostatics, it is not difficult to obtain

$$en = \frac{\gamma \varepsilon}{\kappa} v \frac{\partial T}{\partial x}. \quad (2.6)$$

The pressure is eliminated from the equations of motion after eliminating the charge en . To eliminate the pressure we take the derivative with respect to z of the x -component of the equation of motion, and the derivative with respect to x of the z -component. We obtain

$$v \frac{\partial^3 v}{\partial z^3} - g \beta \frac{\partial T}{\partial x} = \frac{\varepsilon \gamma^2}{\rho \kappa} \left[v \left(\frac{\partial T}{\partial z} \frac{\partial^2 T}{\partial x^2} - \frac{\partial T}{\partial x} \frac{\partial^2 T}{\partial x \partial z} \right) - \frac{\partial v}{\partial z} \left(\frac{\partial T}{\partial x} \right)^2 \right]. \quad (2.7)$$

As noted above, the longitudinal component of the temperature gradient predominates

$$|\partial T / \partial x| \gg |\partial T / \partial z|, \quad (2.8)$$

and the temperature variation along the layer due to the external heat flux is much larger than the temperature variation perpendicular to the layer caused by the motion. This approximation is consistent with our model.

Using this condition, the equation of motion becomes

$$\frac{\partial^3 v}{\partial z^3} - \frac{\beta g}{\nu} \frac{\partial T}{\partial x} = - \frac{\varepsilon \gamma^2}{\rho \kappa \nu} \frac{\partial v}{\partial z} \left(\frac{\partial T}{\partial x} \right)^2. \quad (2.9)$$

On the basis of this equation, the heat conduction equation, and the conditions at the surface and at the bottom of the liquid discussed above, it is shown in the Appendix that $\partial T / \partial x = A$ is constant over the full depth of the liquid, i.e., for $0 \leq z \leq h$. Then the equation of electrostatics (2.6) can be rewritten in the form $en = \gamma \varepsilon A v / \kappa$ and we conclude that in the model considered here the thermoelectric charge is "frozen" into the liquid.

Because the layer is finite along the x axis, the total flow rate of the liquid is zero. Therefore

$$\int_0^h v \, dz = 0. \quad (2.10)$$

In addition, the solution of (2.9) and (2.4) must satisfy certain boundary conditions.

At the solid surface $z=0$ we have the no-slip condition, i.e., $v=0$. The other boundary may be a solid, in which case

$v=0$ at $z=h$, or it may be a free surface, in which the surface tension (the coefficient α) varies because of the heating, i.e., we have the thermocapillary condition

$$\rho \nu \frac{\partial v}{\partial z} = -\sigma \frac{\partial T}{\partial x} = -\sigma A = \text{const}, \quad \sigma = -\frac{\partial \alpha}{\partial T} = \text{const} \quad (2.11)$$

(σ is the thermocapillarity coefficient).

3. EXACT SOLUTIONS

It is convenient to discuss exact solutions of the problem using the dimensionless coordinate $\xi = z/h$ and the Rayleigh number R , Marangoni number M , and the number $\mathcal{E} = I^2$, which characterize, buoyancy, thermocapillarity, and thermoelectricity relative to the dissipative forces:

$$R = \frac{\beta g A h^4}{\kappa \nu}, \quad M = \frac{\sigma A h^2}{\rho \kappa \nu}, \quad \mathcal{E} = I^2 = \frac{\varepsilon \gamma^2 A^2 h^2}{\rho \kappa \nu}. \quad (3.1)$$

Comparing the dimensionless numbers with one another, one can establish a hierarchy of scales.⁶ We have the critical thicknesses

$$h_{RM} = \left(\frac{\sigma}{\beta \rho g} \right)^{1/2}, \quad h_{MI} = \left(\frac{\rho \kappa \nu \varepsilon \gamma^2}{\sigma^2} \right)^{1/2}, \\ h_{RI} = \left(\frac{\kappa \nu \varepsilon \gamma^2}{\beta^2 g^2 \rho} \right)^{1/6}. \quad (3.2)$$

We next write down exact solutions of the problem. When both of the boundaries are solid, we find the following exact solution (independently of the thickness):

$$v = \frac{\kappa}{h} \frac{R}{2I^2} \left\{ 2\xi - 1 + \frac{\cos(I\xi) - \cos(I(1-\xi))}{1 - \cos I} \right\}, \quad (3.3)$$

$$T = Ah \left\{ \frac{x}{h} - \frac{1}{I^2} \frac{v}{\kappa/h} + \frac{R}{2I^2} \left(\frac{1}{3} \xi^3 - \frac{1}{2} \xi^2 + \frac{1}{6} \xi \right) \right\}, \quad (3.4)$$

$$E_z = \gamma A \left\{ \frac{R}{2I^2} \left(\xi^2 - \xi + \frac{1}{6} \right) - \frac{R}{2I^4} \left[2 - \frac{\sin(I\xi) + \sin(I(1-\xi))}{1 - \cos I} \right] \right\}. \quad (3.5)$$

A more realistic situation is the case in which one boundary ($\xi=0$) is solid and the other ($\xi=1$) is a free surface. We consider first a thin film ($h \ll h_{RM}$). Then buoyancy can be neglected ($R=0$), and we find

$$v = \frac{\kappa}{h} \frac{M}{I} \frac{(1 - \cos I)(1 - \cos(I\xi)) - (I - \sin I)\sin(I\xi)}{I \cos I - \sin I}, \quad (3.6)$$

$$T = Ah \left\{ \frac{x}{h} - \frac{1}{I^2} \frac{v}{\kappa/h} + \frac{M}{I^3} \frac{(1 - \cos I)[I^2 \xi^2 + (4 - I^2)\xi] - 2I\xi \sin I}{I \cos I - \sin I} \right\}. \quad (3.7)$$

If the film is sufficiently thick ($h > h_{RM}$), then thermocapillarity can be neglected:

$$v = \frac{\kappa R}{h I^2} \times \left\{ \xi - \frac{[2(1 - \cos I) - I^2 \cos I](\cos(I\xi) - 1) + [2(I - \sin I) - I^2 \sin I] \sin(I\xi)}{2I(I \cos I - \sin I)} \right\}, \quad (3.8)$$

$$T = Ah$$

$$\times \left\{ \frac{x}{h} + \frac{R}{6I^2} \xi \left(\xi^2 - 1 + \frac{6}{I^2} \right) - \frac{1}{I^2} \frac{v}{\kappa/h} + \frac{R}{I^2} \right. \\ \left. \times \frac{[2(1 - \cos I) - I^2 \cos I](1 + I^2 \xi^2) + [8(1 - \cos I) + I^4 \cos I - 4I \sin I] \sin(I\xi)}{4I^3(I \cos I - \sin I)} \right\}. \quad (3.9)$$

The solution for E_z in the presence of a free boundary is quite complicated.

Because of the linearity of (2.9) and the boundary conditions, the solution taking into account both buoyancy and thermocapillarity is simply the sum of the right-hand sides of the equations for the individual forces.

4. ANALYSIS OF THE SOLUTIONS

In the absence of the thermoelectric field, (3.3), (3.4) and (3.6), (3.7) reduce to the exact solutions when only buoyancy or thermocapillarity acts.³ When only thermoelectricity acts, i.e., when $R=0$ and $M=0$ but $I \neq 0$, motion does not occur, i.e., for small heating the motion in the longitudinal direction is produced by buoyancy or thermocapillarity, but not by thermoelectricity.

It can be shown that the electric charge density is the same at the two boundaries,

$$\varepsilon E_z(0) = \varepsilon E_z(h). \quad (4.1)$$

When the temperature at the hot wall increases, the quantity A , and therefore the numbers R , M , and I , increase. An increase in R and M does not change the solution qualitatively, but when I reaches a certain critical value I_* ($I_* = I_r = 2\pi$ for the case of two solid boundaries and $I_* = I_f \approx 4.5$ (the first nonzero solution of the equation $I = \tan I$) for the case when one boundary is free), the solution changes qualitatively. The quantities characterizing the flow increase without bound when $I \rightarrow I_*$, and the component of the thermoelectric force parallel to the layer and the heat flux in the z direction through the surface $z=h$ are

$$F_x = enE_x = \frac{\varepsilon \gamma^2 A^2}{\kappa} v, \quad Q_z = -\rho C_p \kappa \frac{\partial T}{\partial z} \quad (4.2)$$

and the velocity and temperature are both inversely proportional to $I_* - I$. The quantity $F_z = enE_z$ and the heat flux along the layer

$$Q_x = \rho C_p \left(-\kappa \int_0^h \frac{\partial T}{\partial x} dz + \int_0^h v T dz \right) \quad (4.3)$$

are of order $(I_* - I)^{-2}$.

It has been shown that cellular motion occurs when $I = I_*$ in a liquid layer heated perpendicular to the layer.^{7,8} This is due to a thermoelectric force proportional to

$|\nabla T_0|^2 = A^2$, and does not depend on the direction of heating. The values of I_* and I^* necessary for unbounded growth in the case of heating along the layer, and for cellular motion in the case of heating perpendicular to the layer, respectively, are of the same order of magnitude. In the most symmetric form $I_* = I^* = 2\pi$.

This supports the proposition that when the heat flux along the layer reaches the value $I \rightarrow I_*$, flow along the layer is transformed into cellular flow by the thermoelectric mechanism.

Also, when $I \rightarrow I_*$ the condition $v = v_x$ assumed in Sec. 2 is violated, and there is a nonzero component of the velocity v_z perpendicular to the layer.

The quantities $v = v_x$, E_z , and F_z for $I \leq I_*$ are illustrated in Figs. 1–3. Figure 1 shows the dimensionless velocity (curve 1), electric field (curve 2), and force (curve 3) for the case when both boundaries are solid:

$$V_1 = \frac{v}{\kappa/h} \frac{I_r^2(I_r - I)}{R}, \quad E_1 = \frac{E_z}{\gamma A} \frac{I_r^3(I_r - I)}{R},$$

$$F_1 = \frac{F_z}{\varepsilon \gamma^2 A^2/h} \frac{I_r^5(I_r - I)^2}{R^2},$$

(compare with (3.3)–(3.5)).

Figure 2 shows the case when one boundary is solid ($z=0$) and the other is free ($z=h$):

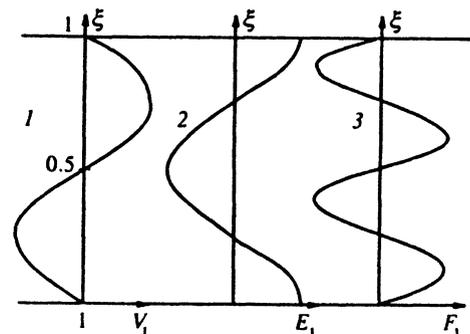


FIG. 1. Distribution of the dimensionless velocity (curve 1), electric field (curve 2), and force (curve 3) in the case of solid boundaries.

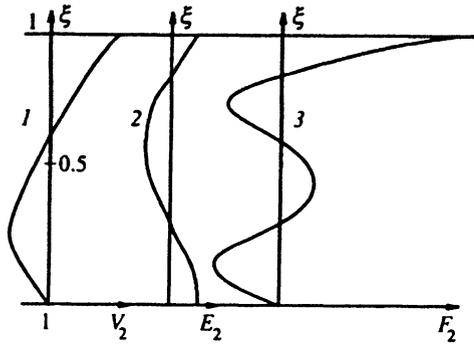


FIG. 2. Distribution of the velocity (curve 1), electric field (curve 2), and force (curve 3) when one boundary ($\xi=0$) is solid and the other ($\xi=1$) is free. Only the thermocapillary and thermoelectric mechanisms are effective; buoyancy forces are neglected (thin layer).

$$V_2 = \frac{v}{\kappa/h} \frac{I_f(I_f - I)}{M}, \quad E_2 = \frac{E_z}{\gamma A} \frac{I_f(I_f - I)}{M},$$

$$F_2 = \frac{F_z}{\epsilon \gamma^2 A^2/h} \frac{I_f^4(I_f - I)^2}{M^2}.$$

In this case we neglect buoyancy in comparison with thermocapillarity, which is valid in thin layers (compare (3.6) and (3.7)).

For the same conditions, but in the absence of thermocapillarity we have plotted in Fig. 3

$$V_3 = \frac{v}{\kappa/h} \frac{I_f^3(I_f - I)}{R}, \quad E_3 = \frac{E_z}{\gamma A} \frac{I_f^5(I_f - I)}{R},$$

$$F_3 = \frac{F_z}{\epsilon \gamma^2 A^2/h} \frac{I_f^8(I_f - I)^2}{R^2}.$$

5. POSSIBLE EXPERIMENTS

The conditions closest to our model are obtained upon alloying steel or semimetal melts (with carbon or tungsten, for example) by means of laser radiation.^{9,10} However the internal flow in such melts has not been studied.

Taking $\beta \approx (9-6) \times 10^{-4} \text{ K}^{-1}$, $\rho \approx 1-10 \text{ g/cm}^3$, $\gamma \approx 10-100 \text{ } \mu\text{V/K}$, $\nu \approx \kappa \approx 5 \times 10^{-2} - 1 \text{ mm}^2/\text{sec}$, $\sigma \approx (1-10) \times 10^{-5} \text{ N/m K}$ for the parameters of the liquid,^{7,8} it can be shown

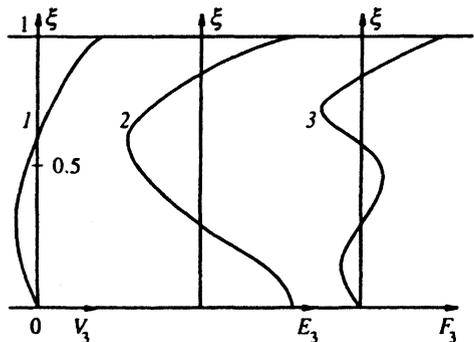


FIG. 3. Distribution of the velocity (curve 1), electric field (curve 2), and force (curve 3) when buoyancy and the thermoelectric force are effective (thick layer).

that for a temperature difference of $\approx 10^3 - 10^4 \text{ K}$ between the center and the sides of the container the dimensionless numbers are such that I is close to I_f . Therefore the distribution of the alloying material over the thickness of the melt will correspond to curve 1 of Fig. 2.

Indeed, in surface laser heating¹¹ the liquid layer is thin, hence the heating is the same over the full thickness. The heated region (the "spot") is large and the temperature difference between the center and the periphery is significant. This situation also corresponds to our model.

6. CONCLUSION

We have shown that in a liquid semiconductor (semimetal) with temperature increasing along the surface, the flow along the layer in fact increases without bound for the same values of the temperature gradient as in the case of cellular motion with heating is perpendicular to the layer. Because the thermoelectric force depends only on $|\nabla T_0|$ and not on the direction of heating, cellular motion also occurs in the case of an external heat flux parallel to the layer.

The author thanks A. M. Bonch-Bruevich and M. N. Libenson for discussions of the experimental situations.

APPENDIX

We consider (2.9) as a quadratic equation in $\partial T/\partial x$. It is evident that because the coefficients of the quadratic equation are functions only of z , the quantity $\partial T/\partial x$ depends only on z . Therefore $\partial^2 T/\partial x^2 = 0$, and the heat transport equation becomes

$$v \frac{\partial T}{\partial x} = \kappa \frac{\partial^2 T}{\partial z^2}. \quad (\text{A1})$$

Differentiating this equation with respect to x , we obtain

$$\frac{\partial^2}{\partial z^2} \frac{\partial T}{\partial x} = 0 \quad (\text{A2})$$

and therefore $\partial T/\partial x = C_1 z + C_2$, and

$$T = (C_1 z + C_2)x + f(z). \quad (\text{A3})$$

The constants C_1 and C_2 and the arbitrary function $f(z)$ are found from the solution and the boundary conditions. We use the fact that in our model the temperature along the bottom and along the surface of the layer varies linearly. We have

$$Ax = C_2 x + f(0) \quad \text{at } z=0 \quad (\text{A4})$$

and

$$Ax = (C_1 h + C_2)x + f(h) \quad \text{at } z=h. \quad (\text{A5})$$

Because these relations must be satisfied for any x , it follows that $C_2 = A$, $f(0) = f(h) = 0$, and hence $C_1 = 0$. Therefore $\partial T/\partial x = C_2 = A$, so $\partial T/\partial x$ is constant for $0 \leq z \leq h$.

¹L. N. Aleksandrov, *Kinetics of Crystallization and Recrystallization of Semiconductor Films* [in Russian], Nauka, Novosibirsk (1985), p. 24.

²S. Yu. Karpov, Yu. V. Koval'chuk, and Yu. V. Pogorel'skiĭ, *Fiz. Tekh. Poluprovodn.* **20**, 1945 (1986) [*Sov. Phys. Semicond.* **20**, 1221 (1986)].

³R. V. Birikh, *Zh. Prikl. Mekh. Tekh. Fiz.* No. 3, 69 (1966).

⁴V. G. Levich and V. S. Krylov, *Ann. Rev. Fluid Mech.* **1**, 293 (1969).

⁵Yu. V. Sanochkin, *Zh. Prikl. Mekh. Tekh. Fiz.* No. 6, 134 (1983).

- ⁶G. Z. Gershuni, E. M. Zhukhovitskiĭ, and A. A. Nepomnyashiĭ, *Stability of Convective Flow* [in Russian], Nauka, Moscow (1989), p. 169.
- ⁷E. D. Eidel'man, *Zh. Éksp. Teor. Fiz.* **103**, 1633 (1993) [*JETP* **76**, 802 (1993)].
- ⁸E. D. Eidel'man, *Zh. Éksp. Teor. Fiz.* **104**, 3058 (1993) [*JETP* **77**, 428 (1993)].
- ⁹N. N. Rykalin, A. A. Uglov, and A. N. Kokora, *Fiz. Khim. Obrab. Mater.* No. 6, 14 (1972).

- ¹⁰A. I. Betaneli, L. P. Danilenko, T. N. Loladze *et al.*, *Fiz. Khim. Obrab. Mater.* No. 6, 22 (1972).
- ¹¹A. G. Grigor'yants and A. N. Safonov, *Methods of Surface Laser Treatment* [in Russian], Vyssh. Shkola, Moscow (1987), p. 136.

Translated by J. D. Parsons